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ENVIRONMENTAL HEALTH SERIES
Radiological Health

**FULL-SCALE SYSTEM FOR REMOVAL OF
RADIOSTRONTIUM FROM MILK**

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

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**FULL-SCALE SYSTEM FOR REMOVAL OF
RADIOSTRONTIUM FROM FLUID MILK**

Proceedings of Seminar
February 24-25, 1965

A cooperation research investigation supported jointly by the U.S. Department of Health, Education, and Welfare, Public Health Service, National Center for Radiological Health, and the U.S. Department of Agriculture, Agricultural Research Service, Eastern Utilization, Research and Development Division.

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

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The Division of Radiological Health was reorganized January 1, 1967. It is now the National Center for Radiological Health, with Mr. James G. Terrill, Jr., as the Director.

Public Health Service Publication No. 999-RH-28

October 1967

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FOREWORD

Since 1956, the Public Health Service and State health agencies have had active programs to assess radioactivity in milk. During the past 5 years, the Public Health Service, the Agricultural Research Service, and the Atomic Energy Commission have been jointly engaged in the development of a method for removal of radiostrontium and other radioactivity from fluid milk. As a result of this work, a full-scale commercial plant was constructed to process milk for strontium removal. The final test of the full-scale plant was scheduled for February 24-25, 1965, during which 200,000 pounds of fluid whole milk were to be processed in a 16-hour period in the fully automated system.

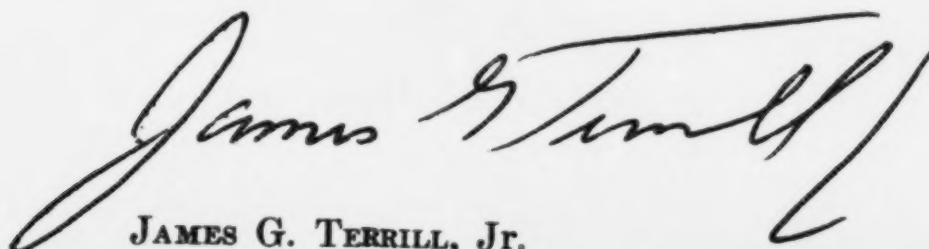
The Public Health Service arranged a seminar to coincide with this final test at the plant site in Lebanon, Mo., so that officials of State health agencies and the milk industry could observe the plant in operation and afterward discuss the process as it related to their fields of interest. The Department of Agriculture was asked to participate in the seminar and to invite agricultural groups who might also contribute.

This combined demonstration and seminar was considered to be the best way to foster an exchange of views on the process among representatives of health, agriculture, and industry, and to obtain their comments on technical limitations in the process which would work to the disadvantage of the milk industry or Government agencies during emergency periods.

Participants were asked to submit comments after the meeting on the basis of their observations. As a followup to the seminar, it was planned to provide participants with a record of the proceedings and with information on the economics of the process.

The success of the seminar was due in a large measure to the planning of the Project Director, Mr. Jack H. Fooks, the conscientious efforts of Mr. Ronald E. Bales and his staff, the full cooperation of Dr. Sam R. Hoover, Mr. Thomas W. Quigley, and Mr. Homer E. Walter of the Agricultural Research Service, and to the other individuals who

contributed the papers recorded here. The manuscript was prepared by Mrs. Annette Urciolo and Mrs. Joyce Merry of the Research Branch, Division of Radiological Health.

A handwritten signature in cursive script, reading "James G. Terrill, Jr.", with a long, sweeping underline.

JAMES G. TERRILL, Jr.
Director, National Center for Radiological Health
Public Health Service
Department of Health, Education, and Welfare

OCTOBER 1967

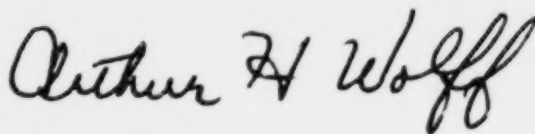
PREFACE

Since 1960, the Public Health Service, the Agricultural Research Service, and the Atomic Energy Commission have jointly engaged in the development of a method for removal of radiostrontium and other radioactivity from fluid milk by the ion exchange method. These investigations progressed in a series of evaluations from the laboratory through the pilot plant, culminating in a full-scale commercial process developed under a contract supported jointly by the Department of Agriculture and the Department of Health, Education, and Welfare, with the Producers Creamery Company of Springfield, Mo.

As a part of the contract, the contractor has constructed a full-sized cation-resin strontium removal plant at Lebanon, Mo. During its evaluation, the plant processed over 500,000 pounds of fluid whole milk, and 300,000 pounds of skim milk for strontium removal.

This seminar was held in Lebanon on February 24-25, 1965, to coincide with the final test of the treatment plant, and included presentation of papers covering various aspects of the investigations and plans for future research and development as well as observation of the treatment system in operation. Seventy-one representatives of State health agencies, State agricultural agencies, the Public Health Service, the Agricultural Research Service of the U.S. Department of Agriculture, the Canadian Department of Agriculture, and key personnel of the milk industry participated.

These proceedings were prepared from the records of the seminar to provide information for members of the scientific community and industry, and to document the progress and accomplishments of the project. A list of participants and the agenda for the 2-day session are included as a part of the record.



Arthur H. Wolff
Chief, Radiation Bio-Effects Program
National Center for Radiological Health
Public Health Service
Department of Health, Education, and Welfare

OCTOBER 1967

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ABSTRACT

A seminar was held simultaneously with a demonstration of a full-scale system for removing radiostrontium from fluid milk at the Producers Creamery Company, Lebanon, Mo. The removal rate of the process was better than 90 percent. The taste of the treated milk compared favorably with regular milk. Nutritional values were altered somewhat, but animal feeding studies showed no deleterious effects on growth or general health as a result of known changes in the processed milk. The system included storage tanks, ion exchange resin columns, a high-temperature short-time pasteurizer, time and step controllers, and pH controls. Papers were presented on laboratory studies of ion exchange methods, nutritional investigation of milk processed in a pilot plant, microbiological aspects of the fixed-bed process, design and operation of full-scale equipment, problems associated with designing equipment, automation of full-scale equipment, problems associated with quality control and removal efficiency, relationship of the ion exchange process to present day commercial fluid milk processing operations, and additional research and development activities considered for treatment processes.

Representative products and manufacturers are named for identification only, and listing does not imply endorsement by the Public Health Service and the U.S. Department of Health, Education, and Welfare.

Prime Contractor :

**Producers Creamery Company
Springfield, Mo.**

Plant Location :

Lebanon, Mo.

Project Director :

**Jack H. Fooks, Research Branch
Division of Radiological Health
Public Health Service, DHEW**

Contracting Officer's Designated Representative :

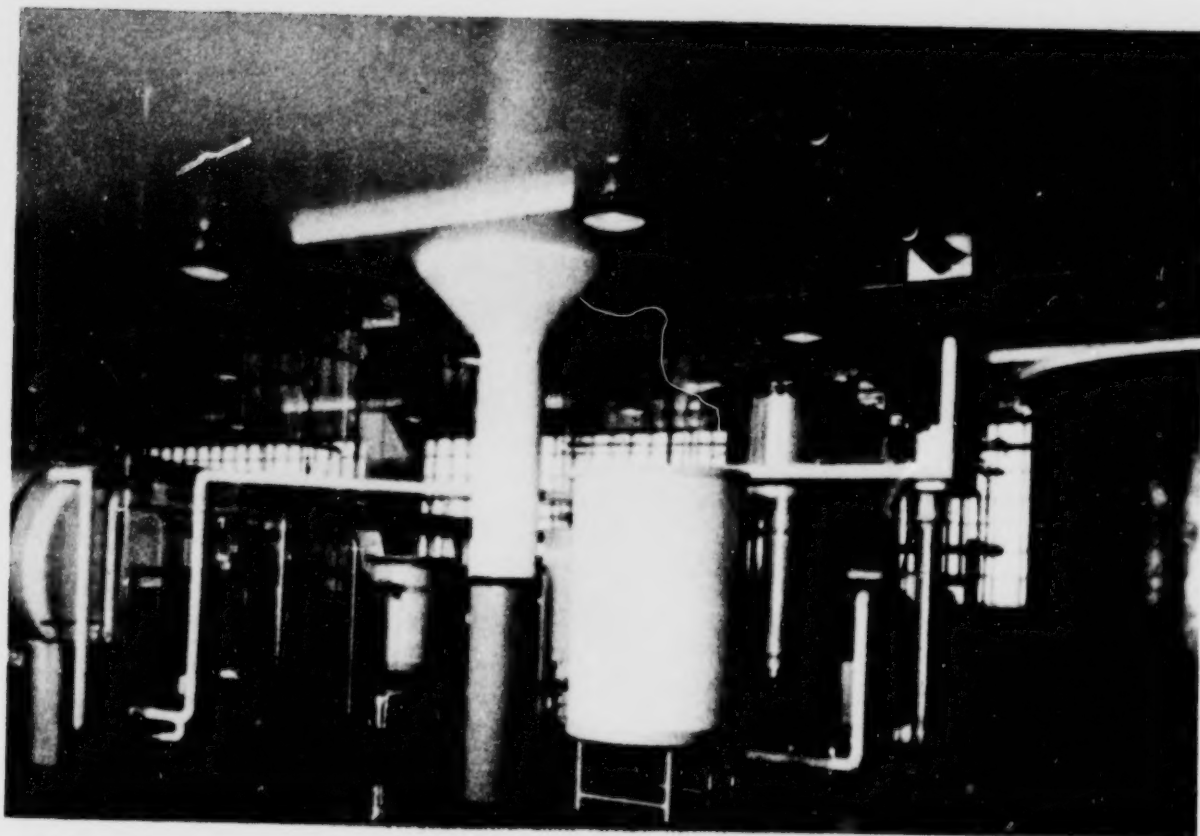
**Homer E. Walter
Eastern Utilization
Research and Development Service
U.S. Department of Agriculture**

Joint Advisory Committee for Removal of Radiostrontium from Milk :

**Chairman : Sam R. Hoover
Agricultural Research Service
U.S. Department of Agriculture**

**Cochairman : James G. Terrill, Jr.
Division of Radiological Health
Public Health Service, DHEW**

**Executive Secretary : Thomas W. Quigley
Agricultural Research Service
U.S. Department of Agriculture**



The equipment pictured in the above photograph is the actual commercial-scale, fixed-bed system for removal of radiostrontium from fluid milk installed at the Producers Creamery Company plant in Lebanon, Mo. Operation of the equipment has successfully demonstrated the feasibility of using this ion exchange system as an emergency treatment process for removal of radioactivity in a commercial milk plant.

ATTENDEES ¹

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¹ Affiliations and addresses of attendees as of the date of the symposium.

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Rockville, Md.

John O. Young
Purdue University
Lafayette, Ind.

Division of Environmental Engineering and Food Protection
Public Health Service
Washington, D.C.

3:40 p.m.-3:50 p.m.

Discussion

3:50 p.m.-4:10 p.m.

Additional Research and Development Activities Considered for Treatment Processes

Mr. R. E. Bales, Chief, Environmental Contract Research, Research Branch, Division of Radiological Health, Public Health Service, Rockville, Md.

4:10 p.m.-5:00 p.m.

Summary Discussion

February 25

9:00 a.m.

Observation of Regeneration and Precharging Cycle for the Full-Scale Plant and Observation of 16-Hour Fluid Milk Treatment Test at Producers Creamery Company Plant
Lebanon, Mo.

Mr. H. Hurst, Plant Manager, Producers Creamery Company
Lebanon, Mo.

AGENDA

SEMINAR ON THE FULL-SCALE SYSTEM FOR REMOVAL OF RADIOSTRONTIUM FROM FLUID MILK

Lebanon, Mo., February 24-25, 1965

Morning Session—February 24

Chairman: Mr. James G. Terrill, Jr.
Deputy Chief, Division of Radiological Health
Public Health Service
Washington, D.C.

8:30 a.m.—8:45 a.m.

Introductory Remarks

Dr. Sam R. Hoover
Staff Scientist
Research Program Development and Evaluation Staff
Washington, D.C.

8:45 a.m.—9:00 a.m.

Historical Background

Mr. J. H. Fooks, Project Director
Division of Radiological Health
Public Health Service
Division of Radiological Health
Rockville, Md.

9:00 a.m.—9:20 a.m.

Preliminary Laboratory Studies on Ion Exchange Processes for Treatment of Fluid Milk

Dr. G. K. Murthy, Milk and Food Research Activities
Robert A. Taft Sanitary Engineering Center
Division of Environmental Engineering and Food Protection
Public Health Service
Cincinnati, Ohio

9:20 a.m.-9:30 a.m.

Discussion

9:30 a.m.-9:50 a.m.

The Beltsville Process

Dr. L. F. Edmondson, Project Leader
Radioisotopes Removal Project
Dairy Products Laboratory
Eastern Utilization Research and Development Division
Agricultural Research Service
Beltsville, Md.

9:50 a.m.-10:00 a.m.

Discussion

10:00 a.m.-10:30 a.m.

Movie: The Removal of Radiostrontium From Milk

10:30 a.m.-10:40 a.m. Break

10:40 a.m.-11:00 a.m.

Nutritional Investigation of Pilot Plant Processed Milk

Dr. D. G. Hazzard, Biochemist, Division of Radiological
Health, Public Health Service
Division of Radiological Health
Rockville, Md.

11:00 a.m.-11:10 a.m.

Discussion

11:10 a.m.-11:30 a.m.

Sanitary Aspects of Beltsville Process

**Dr. R. B. Read, Jr., Milk and Food Research Activities
Robert A. Taft Sanitary Engineering Center
Public Health Service
Cincinnati, Ohio**

11:30 a.m.-11:40 a.m.

Discussion

12:00 m.-1:00 p.m. Lunch

Afternoon Session

**Chairman: Dr. F. A. Todd, Assistant to Administrator
Agricultural Research Service
U.S. Department of Agriculture
Washington, D.C.**

1:00 p.m.-1:20 p.m.

Design and Operation of Full-Scale Equipment

**Mr. H. E. Walter, Dairy Technologist and Contracting
Officer's Representative, Radioisotope Removal Project,
Dairy Products Laboratory, Eastern Utilization Research
and**

**Development Division, Agricultural Research Service
Beltsville, Md.**

1:20 p.m.-1:30 p.m.

Discussion

1:30 p.m.-1:50 p.m.

Problems Associated with Equipment Design

**Mr. J. H. McCutchen, Sanitary Engineer
Special Projects Section, Milk and Food Branch
Division of Environmental Engineering and Food Protection
Public Health Service
Washington, D.C.**

and

Mr. D. E. Body,¹ Mechanical Engineer
Aerometry Section, Field Studies Branch
Air Pollution Research Activities
Robert A. Taft Sanitary Engineering Center
Division of Air Pollution, Public Health Service
Cincinnati, Ohio

1:50 p.m.-2:00 p.m.

Discussion

2:00 p.m.-2:20 p.m.

Automation of Full-Scale System

Mr. G. Sparling, Supervisor of Sanitation and Quality
Control
Producers Creamery Company
Springfield, Mo.

2:20 p.m.-2:30 p.m.

Discussion

2:30 p.m.-2:50 p.m.

Problems Associated with Quality Control and Removal
Efficiency and Problems Associated with Processing

Mr. B. H. Heinemann, Technical Manager
Producer Creamery Company
Springfield, Mo.

2:50 p.m.-3:00 p.m.

Discussion

3:00 p.m.-3:20 p.m. Break

3:20 p.m.-3:40 p.m.

Relationship of the Ion Exchange Process to Present Day
Commercial Fluid Milk Processing Operations

Mr. E. L. Ruppert, Chief, Milk and Food Branch

¹ Formerly Field Project Engineer, Radiological Health Laboratory, DRH,
PHS, Rockville, Md.

FULL-SCALE SYSTEM FOR REMOVAL OF RADIOSTRONTIUM FROM MILK

INTRODUCTORY REMARKS

Sam R. Hoover¹

Mr. J. G. Terrill, Jr.:

A compromise between our schedule and the weather seems indicated. Perhaps even starting now is a little bit unfair, but I think if we wait too long it will be unfair to the rest of you, so even at the risk of seeming unreasonable, we will go ahead with the meeting.

I want to welcome all of you on behalf of not only our Department but of the joint group that has worked on this project for a number of years. I also want to mention very briefly the purpose of the meeting. The meeting is not an information-type meeting in the usual sense, but rather an opportunity for us to present what people in the Producers Creamery Company, the Department of Agriculture, and our agency have done up to this point to try to develop a practical method of removing strontium 90 from milk. We hope to get your views, just as we have those of our laboratory and technical people, on how far along we are in meeting our original objectives and where we go from here. We understand that this is a closed meeting. There will be no effort to entertain reporters, so you can speak with that knowledge in mind.

To open the program today, we've asked Dr. Sam R. Hoover to make introductory remarks. I might say, in introducing Dr. Hoover, that he has been the senior Department of Agriculture representative on the joint, interagency planning group for the project. It has been a pleasure to work with him over the years, and he has proven to be a very constructive colleague in this endeavor.

¹ Research Program Development and Evaluation Staff, Agricultural Research Service, U.S. Department of Agriculture.

Dr. Hoover:

I have a somewhat difficult job here not to interfere with what Mr. Fooks is going to say because he is to give a background statement, and one does not start off in the first talk of the meeting by offering everything that will be said if one can avoid it.

The beginning of a venture is difficult to pinpoint in many cases. You can, if you wish, say that the first ideas on removal of strontium 90 from milk began with the concept of Democritus that there were atoms. However, you can place a definite beginning on this particular venture and although it might appear somewhat immodest I am going to place it quite directly.

I was having a gin and tonic with my wife in the backyard after dinner on Wednesday, April 29, 1959. As usual, I was talking about things that people should do that they did not do, and so forth, and I spoke of this problem. And, with the sort of single-minded logic that women sometimes have, she said, "Well, if there is anybody in a position to do something about this, it is you." So we talked a bit, and the next morning I went to the Department of Agriculture and wrote a memorandum. I brought it to read to you.

"To: Dr. George W. Irving, Jr.
Deputy Administrator

Subject: Decontamination of Milk

"The levels of radioactive strontium 90 (^{90}Sr) in milk are increasing, and it has been authoritatively stated that the amount of fallout will increase over the next ten years two- to three-fold from the strontium 90 already in the upper atmosphere. Inasmuch as milk is the major way in which strontium 90 gets into the human system, these facts constitute an extremely severe threat to the dairy industry in the United States. Worse than that, it constitutes a threat to the health of the entire milk-drinking population.

"The permissible level of strontium 90 in milk is a moot point. Various levels have been proposed by different agencies. All of them appear to be the merest of guesses. However, values of 20 and 80 S.U. (micro-microcuries per liter) are in use. The Public Health Service shows values of 4 to 18 in various portions of the country in the attached release of Monday, April 13.

"I, therefore, recommend strongly that a 'crash' program be instituted to develop the most economically feasible means of removal of strontium 90 from milk. This will have to be done on a pilot plant and

commercial basis. It will require substantial sums of money. We have suitable space in our pilot plant at Beltsville which can be assigned to such a project. It is not necessarily a laboratory research venture. There is information available upon which pilot-plant investigations could be begun as soon as the necessary equipment and personnel were made available. It is, therefore, well adapted to a crash program attack, one that we should institute as soon as possible."

Dr. Irving took this memorandum to Dr. Shaw, Administrator of the Agricultural Research Service, and by a remarkable set of circumstances, it was in the hands of Assistant Secretary Peterson that afternoon. On Friday morning word came back that the Department of Agriculture would be pleased to authorize us to go ahead with one proviso, and this Dr. Shaw made quite clear. The Public Health Service and the Atomic Energy Commission had to be in the picture, and as partners.

I have read this memorandum to you because I am rather proud of it. What I said there has turned out to be remarkably accurate.

The work here, which is a major subject of discussion, is a test of feasibility. Feasibility is a big word and there will be more discussion of it as we go along. There is no question in my mind that when people approaching the competence of the Producers Creamery personnel do the work, the process is certainly operable. It has also been shown that this design, based on the best estimates of 2 years ago, is not a final design, but we are pleased to give you a progress report up to where we stand now. The economic evaluation is a major part of this work, and until the work is completed and economists have been able to study the data, we do not have all of the answers.

Another point I want to make, and Jim Terrill referred to it, is that there has not been very much red tape, especially for a tripartite job done by three agencies of government. We are justifiably proud of this aspect. As some people here well know, things can get pretty complicated in just one agency of government.

This is not the first public report we have given on the work. On December 19, 1961, pilot-plant work at Beltsville, Md., had gone far enough along, based on the laboratory work at Cincinnati and the work of other people too, so that we announced to the industry what we were doing, why we were doing it, and how far we had gotten along with the job. There is not a great deal of change in concept from that time until now, but there has been a lot of combined work put into this venture. I am very pleased to give you this introduction, and we will learn a lot more as the day goes on.

HISTORICAL BACKGROUND

*Jack H. Fooks*¹

One of the things that Dr. Hoover did not say is that most of this work has been done through the cooperation of technical people to a greater degree than through administrative people. The reason the red tape has been cut to a minimum, I think, is that the technical people actually carried the ball, worked as a team, and ended up as a real producing group for output of technical information. In this folder which we have tried to put together, containing as many of the reprints as we thought were appropriate, you'll find that there has been a considerable amount of published material on this process, both in the laboratory stage and the pilot stage, and we expect that as a result of this meeting there will be some more published material, at least of a preliminary nature, on the output from the full-scale evaluation. Before I begin discussing the historical background I might point out a few things that I would like you to observe when you go down to the plant. As you will recognize, we have been fortunate in becoming involved with a commercial group who have permitted us to use their facilities to evaluate this system. In the plant they are also doing research on processes which have nothing to do with the system for removal of radioactivity, and because of this they have limited the areas where they will permit us to go when we attend the demonstration tomorrow. I am requesting that all of you observe the signs that say "authorized personnel only" and not try to go into that area of the plant.

Another request is not to take pictures. In the folder you will find a complete set of pictures—reprints that we made from pictures that have already been taken. I think they are excellent for the purpose of giving you an idea of what the plant is like, its size, and other schematic diagrams. There is no need to take additional pictures unless it might be of the group attending the meeting, and if you want to do this in the snow, then they will be outside the plant.

¹ Research Branch, Division of Radiological Health, Public Health Service, U.S. Department of Health, Education, and Welfare. Present assignment: Sterility Control Office, National Aeronautics and Space Administration, Washington, D.C.

In getting to this subject of historical background, I think one of the things that might be brought out is that this process is not unique; it has been known for some time. It has been used previously by other food processors in the wine industry, the sugar industry, and even in the milk industry in some of the low-sodium milk applications. The unique part of this has been the development of the system involving the regeneration step, the acidification step, and the precharging of the resin. This laboratory research, of course, has been done in a good number of places by many investigators at the locations where this work began. I think the Canadians should be given credit for being first; namely, Migicovsky and his group in the Canadian Department of Agriculture. Dr. Murthy, before he came to the Sanitary Engineering Center and while at the University of Illinois, did some early work on this. Dr. Edmondson experimented in California with the technique, and quite a number of others have had some input as you can see from the reprints. I might also point out that the actual first use of ion exchange resins for removal of radioactivity from milk was not done by any of the investigators normally associated with milk technology. The University of California Radiation Laboratory, in 1954, as a contractor for AEC demonstrated that it was possible to remove approximately 90 percent of the calcium and strontium from milk using an organic resin. This was explored further under an AEC contract with the University of Tennessee, and further studies were done at the Robert A. Taft Sanitary Engineering Center as well as in England and Canada. From this, you realize that many people have had an input into the system, and the Department of Agriculture was not alone in its efforts to reduce the strontium level in fluid milk.

As Dr. Hoover has pointed out, during the latter part of 1959 the Department of Agriculture approached the Public Health Service and the Atomic Energy Commission with a plan for a cooperative project. This was proposed as a pilot-plant scale plan and was to direct specific attention to the problems of flavor, nutrition, sanitation, and toxicity. In addition, of course, was the need to determine how to improve the removal efficiency, not only for radiostrontium but for other radionuclides in the milk. This plan was divided into three phases: the first was to determine the effectiveness of the various resins that were available on the removal efficiency for radiostrontium; the second was to determine what nutritional and organoleptic changes may occur in the milk, and the third phase (assuming that the first two phases were accomplished) was to develop a pilot plant capable of treating 100 gallons of milk per hour. In September 1959, at the suggestion of the Public Health Service, a joint advisory committee for overall planning was established with membership from

the three Federal agencies and the Canadian Department of Agriculture. The resultant organization was not only a government inter-agency group but an international one. This committee, although called advisory, actually has been a steering committee, and although Dr. Hoover glossed over this point, I feel the reason we have had a successful administrative enterprise has been the fact that the projects have been operated on a membership basis. No unilateral action has been taken without the approval of the other agencies involved. This has reduced many administrative problems although there have been difficulties at times when differences of opinions arose.

In July 1960, a public patent was secured on the process after 6 months of laboratory and pilot-plant work. This patent was taken out by the Department of Agriculture as a public patent in the names of Murthy, Campbell, Masurovsky, and Edmondson. I believe, since Drs. Murthy and Edmondson are both here, this can be verified. This patent refers to the specific acidification step of the process rather than the ion exchange system. The ion exchange portion was not patentable because it was not unique to the process and only the acidification step was considered an innovation.

During the following year, investigations in the laboratory and work on the pilot plant concentrated on the effect of passing the milk through the fixed-bed, cation exchange columns and were related to the amount of strontium removed, changes in cation composition of the treated milk, and flavor changes. The success in this effort led to nutritional studies which Dr. Hazzard will cover. These were begun in the latter part of 1961 and were designed to evaluate the effect of the fixed-bed cation process on treated milk. At the same time, studies were also initiated at Beltsville on the economics of the fixed-bed system and a subcommittee was appointed by the advisory committee to consider the problem of developing a commercially feasible system. On September 6, 1962, the joint advisory committee recommended that the agencies consider going ahead with a commercial venture. It should be noted that the Atomic Energy Commission did not believe they should support the development of the full-scale operational plant because the practical application of the research was not within their charter. Therefore, they did not join in this part of the venture but remained as a member of the advisory group and have continued in this capacity.

You may wonder on what basis Producers Creamery Company was selected as a contractor. The Public Health Service had already had dealings with Producers Creamery on another project which is not associated directly with a plant operation but with a farm operation.

However, the advisory committee considered many other plants with the idea that a suitable place could be found to do this evaluation. Producers Creamery Company was selected on the basis that it was not in the retail milk business, could not derive an economic advantage from exploitation of the publicity resulting from this research, and was in a milkshed area where above normal strontium 90 levels occur in milk. By above normal I do not mean they are excessively above the normal for the United States but are considered high enough that when you remove 90 percent of the strontium 90, enough is left in the milk to be measured with reasonable accuracy. This is critical for evaluation of removal efficiency. As you know, these chemical determinations for radiostrontium are not developed to the *n*th degree, and sensitivity for strontium is roughly around 3 picocuries per liter. Thus, if reduced concentrations in the milk are 10 percent of the original value, it is possible to have a very small number in the final product. The final consideration in the selection was the need to be in an area where diversion of milk from the shed into this process did not upset the economic market (and it isn't a small diversion; 100,000–200,000 pounds of milk a day is considerable). One of the reasons you are sitting here today rather than some other day of the week, as Homer Walter points out, is because Sundays and Wednesdays are the best days to divert milk from the milkshed from the producer's point of view.

The final administrative development in this cooperative program was the generation of a Memorandum of Understanding between the Departments of Agriculture and Health, Education, and Welfare in October 1962 which set up the rules—if you want to call them that—relative to each agency's responsibilities in this cooperative enterprise. Specifically, the Agricultural Research Service was empowered to negotiate a contract by designating a contracting officer's representative (Mr. Homer Walter), and to develop jointly with the Public Health Service general specifications for the prospective contractor's guidance. In turn, the Public Health Service was to provide the project leader, assist in the development of the contract document and specifications, and assume overall responsibility for the conduct of the project. The working draft of the contract document was approved by both agencies and an exploratory visit was made to Producers Creamery Company during September 1962. It should be pointed out that it was not without reluctance that Producers Creamery Company engaged in this contract because they realized that they were in a vulnerable position from the point of view of the other members of the industry, and therefore the Company had to submit this

proposal to their Board of Directors. The Board's decision was favorable and on October 1 Producers Creamery Company informed the Government that they were willing to enter into a contract. The following few months were spent in technical discussions with the Company and other personnel relative to the type of plant that was to be designed. Personnel of the contractor visited Beltsville as well as the proposed equipment fabricators, and the entire development of the full-scale system with the hardware that went with the process actually was a cooperative enterprise from the very beginning. The Government people realized that both Paul Mueller Company, the fabricators of the equipment, and Producers Creamery Company had an excellent staff to do the work and, again as Dr. Hoover pointed out, we, as a Government group, stayed out of the very minute details of the process development so as to give the contractor the freedom necessary to develop a workable system. Again, we respected the technical competence of Producers and Mueller and believed that we would get the job done faster if the Government did not impose its ideas upon them.

In March 1963, an estimate of the cost, based upon the testing schedule which the Government had proposed, was submitted to the joint advisory committee. This shocked quite a few people because the amount of money was roughly three times that which either agency had available. Following this, renegotiation with the Company and modification of the extent of testing produced an agreeable price. Roughly, we had to cut everything down by a factor of 2 and we ended up with a total contract price for this enterprise of \$364,000.

On May 22, the first official test milk run was conducted at the plant in Lebanon and involved 100,000 pounds of skim milk.

I would like to point out that the papers to be presented here will report not only on the laboratory investigations and the pilot-plant work, but the Producers Creamery staff will also present information on the results thus far of the full-scale system.

REMARKS

Mr. Terrill:

Thank you, Jack. I guess we have most of the people here who are going to show up this morning so perhaps it would be appropriate at this time to ask each of you to identify yourselves for the benefit of the rest of the group, and we will also ask Mrs. Smith if she will pass papers around to supply your name and affiliation.

One thing that I think we should mention (I try to keep notes on pertinent points that might have been missed by Sam Hoover or Jack Fooks) is the question of when this process should be used.

This program was set up with the idea of creating a system that could be used if needed and this is as far as we have carried it at this time, and it may be as far as we ever carry it. The question is, "what is an emergency"? This particular point is under consideration by the Federal Radiation Council. We, at this meeting, do not wish to influence the judgment of the Council one way or the other, but to show the process to this cross section of people who represent the agricultural interests, the processing interests, the consumer interests, and the public health interests, and to make available to them not only the Federal Radiation Council's judgment of a process that might be satisfactory in an emergency, but some of your thoughts and conclusions on how we should define an emergency under these kinds of circumstances. However, this particular meeting is only to develop further information that the council might use rather than to try to arrive at a judgment.

Now, at this time, we enter a phase of our program that is related to the laboratory research aspects of the process. We will have some brief comments by Dr. Murthy of the Public Health Service and Dr. Edmondson of the Department of Agriculture who, as Jack Fooks indicated, have worked together very closely. We are going to call on Dr. Murthy first to give us his presentation.

LABORATORY STUDIES ON THE ION EXCHANGE METHODS FOR THE REMOVAL OF RADIONUCLIDES FROM FLUID MILK

*G. K. Murthy*¹

As you know, the Milk and Food Section of the Division of Environmental Engineering and Food Protection of the Robert A. Taft Sanitary Engineering Center in Cincinnati has been engaged in radiation surveillance of milk since May 1957. Analysis of milk samples for radionuclides collected from different geographical locations in the United States indicated a steady increase with time of the concentration of strontium 90. The succession of tests of nuclear weapons by various countries, including installation of nuclear reactor facilities in the United States, indicated that the level of radioactive contamination may increase to an extent that might be deleterious to health, particularly of children. It was obvious to us then that apart from routine surveillance work and research in developing simplified methods for detecting radionuclides in milk, we should also be looking into the matter of decreasing radioactive contamination to safe levels if the occasion warranted it.

There were two approaches to the problem: (1) reduction of the concentration at the point of production on the dairy farm by modification of farm practices, and (2) treatment of milk at the processing plant before the milk reached the consumer. Since the first approach was not suitable for laboratory investigation, consideration was given to the second approach.

A survey of the literature indicated that several investigations had been made to remove strontium, cesium, and iodine from milk by various means, including the ion exchange process. Demott and his associates at the University of Tennessee used ion exchange resins in the calcium, sodium, and potassium cycle to remove strontium and cesium from milk. Although they were able to remove 80 to 90 per-

¹ Robert A. Taft Sanitary Engineering Center, Public Health Service, U.S. Department of Health, Education, and Welfare. At present Dr. Murthy is research chemist of the National Center for Urban and Industrial Health, USPHS, DHEW, 222 East Central Parkway, Cincinnati, Ohio 45202.

cent of the strontium 90, the milk was drastically altered in some of its physical and chemical properties. I think that Migicovsky of Canada was the first person to conceive the idea of charging the resin with a solution containing calcium, sodium, and potassium in approximately the same proportions as it occurred in milk, so that when milk was treated the cationic composition remained unaltered. However, the removal of strontium was rather poor, only 70 percent. Realizing that the protein-bound radionuclides were not available for ion exchange, he thought of using resins charged in two ways. In the first treatment, calcium and strontium both were removed; in the second, the resin gave up calcium and removed excess sodium from milk. This procedure removed about 85 percent of the strontium. All this work was done by the batch technique, and therefore was not suitable for column techniques.

From these observations it was obvious that we had to devise a technique for continuous operation whereby 95 percent of strontium may be removed without any significant changes in the cationic composition of milk. It was apparent that the removal of radionuclides from milk could be achieved only if the protein-bound ions were transformed into more readily exchangeable forms. The physicochemical nature of milk is such that alteration of its pH from 6.6 to 5.2 by some means will shift the equilibrium so that the colloiddally bound ions are released to become free ionic species. To ionize protein-bound salts, acids like citric, lactic, and hydrochloric were considered. Citric acid was the logical choice because it is a natural constituent of milk and is a food additive.

For all investigations, milk, labeled *in vitro* was prepared by adding an aliquot of strontium 85, barium 140, or cesium 134 of known activity to milk and allowing it to equilibrate at 4° C. for 72 hours. The resin used was Dowex-50W-X8 (H⁺), 20-50 mesh size. The resin was charged with a solution containing calcium, magnesium, sodium, and potassium in the same relative proportions as in the milk but in 5 times the concentration, and the pH was adjusted to 5.4. Prior to ion exchange treatment, milk was acidified to pH 5.3 to 5.4 with 0.5M citric acid.

Effect of acidification of milk on the removal of radionuclides from milk

The divalent cations (such as calcium, magnesium, strontium, and barium) are distributed between the serum and colloidal phases. Acidification of milk ionizes the colloiddally bound salts. Figure 1 shows that the removal of strontium 85 and barium 140 are affected by the degree of acidification, the removal dropping from 90 to 53 percent of strontium 85 and 94 to 23 percent of barium 140, as pH was increased

from 5.2 to 6.6. Removal of more strontium 85 than barium 140 at any given pH suggests a more tenacious binding of barium than of strontium in the colloidal protein phases of milk. To remove a high percentage of strontium and barium it is necessary to reduce the pH to 5.3 to 5.4. When lowered to 5.2, the pH becomes critical to protein stability.

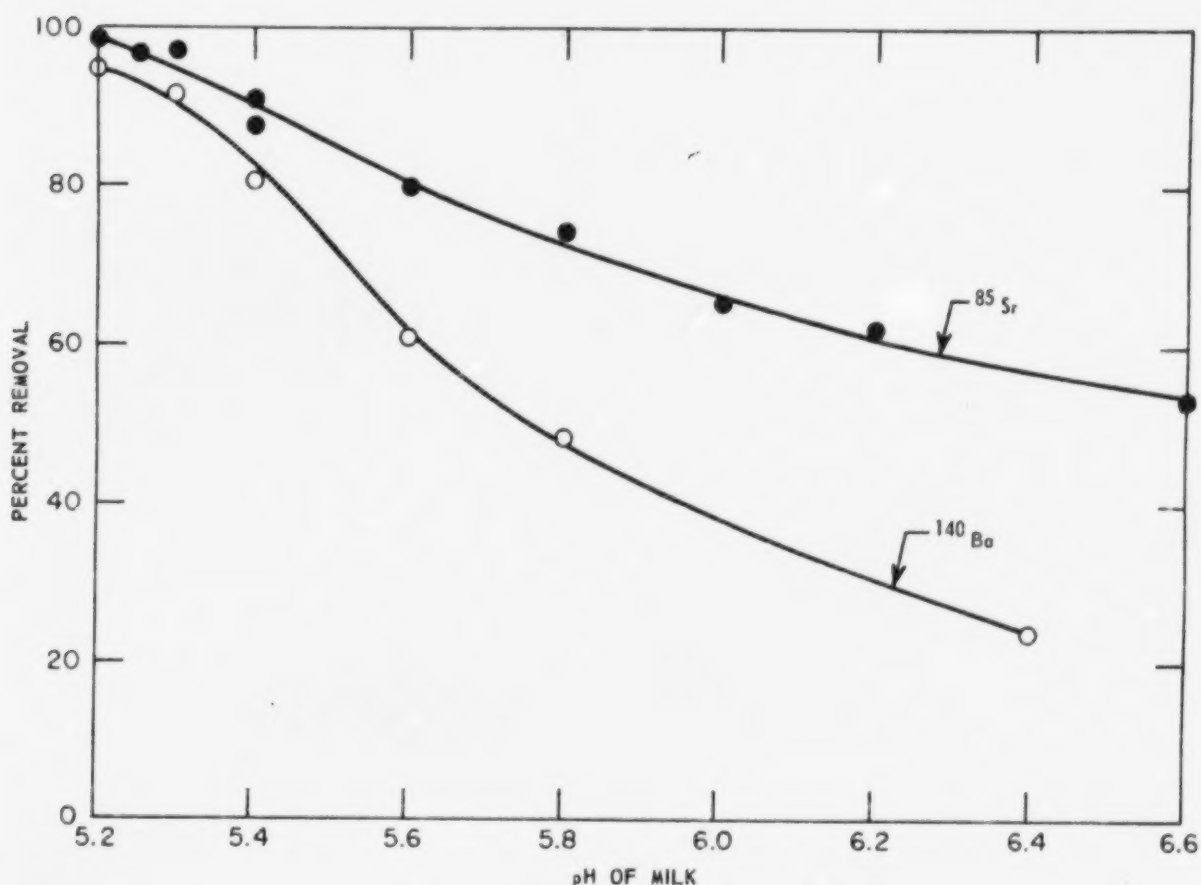


Figure 1. Effect of acidification of milk with citric acid on the removal of ⁸⁵Sr and ¹⁴⁰Ba from milk

Figure 2 shows the removal of cesium 134. The removal of cesium is not affected by acidification of milk. This is to be expected, since monovalent cations are in the free ionic state in milk. However, the cesium 134 is not removed as efficiently as strontium 85 or barium 140 because of preferential absorption of divalent rather than monovalent cationic radionuclides.

Comparison of strontium 85 removal from milk labeled in vivo and in vitro

Because of the differences observed by other investigators in the removal of strontium from milk labeled *in vivo* and *in vitro*, it was important to establish whether any significant differences existed in the removal of strontium when milk was processed at pH 5.3 to 5.4. Figure 3 shows that at pH 5.2 to 5.3 approximately 93 percent of strontium

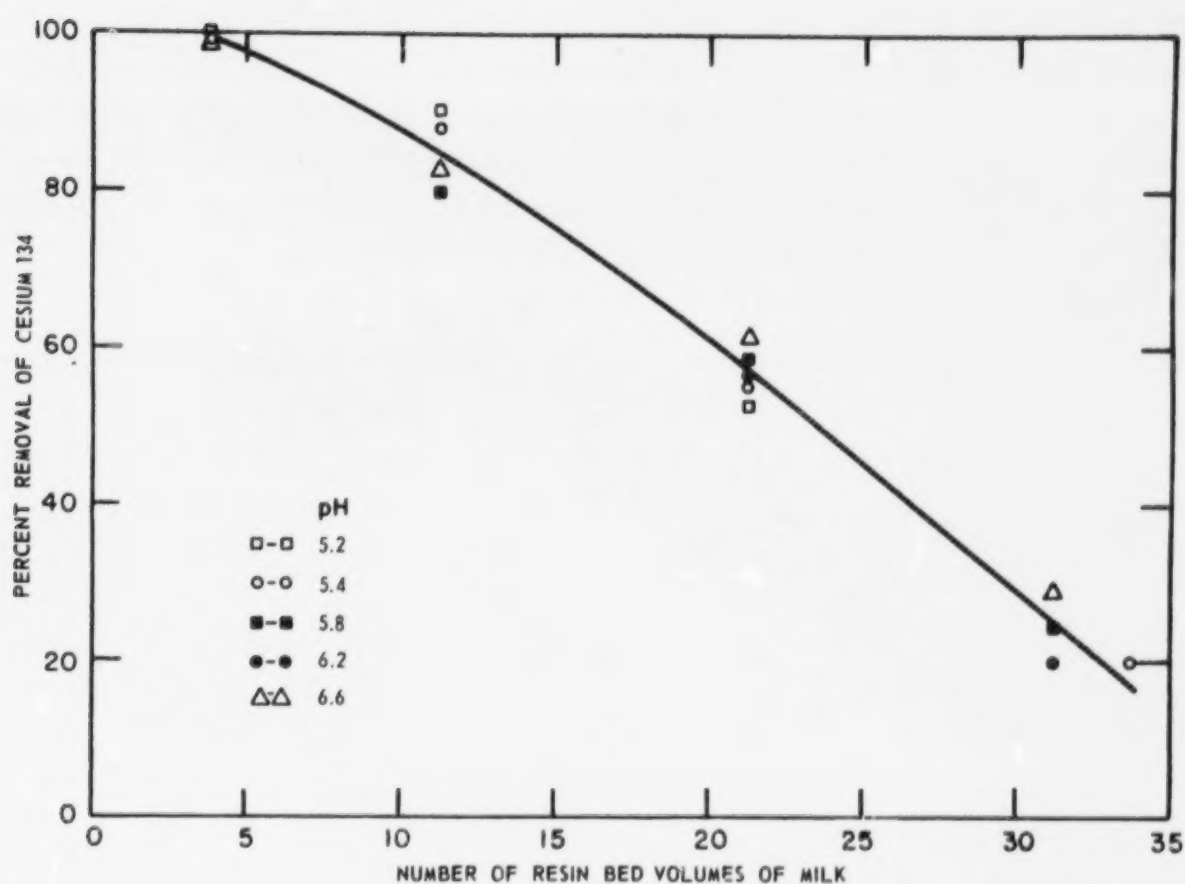


Figure 2. Effect of acidification of milk with citric acid on the removal of ^{134}Cs from milk

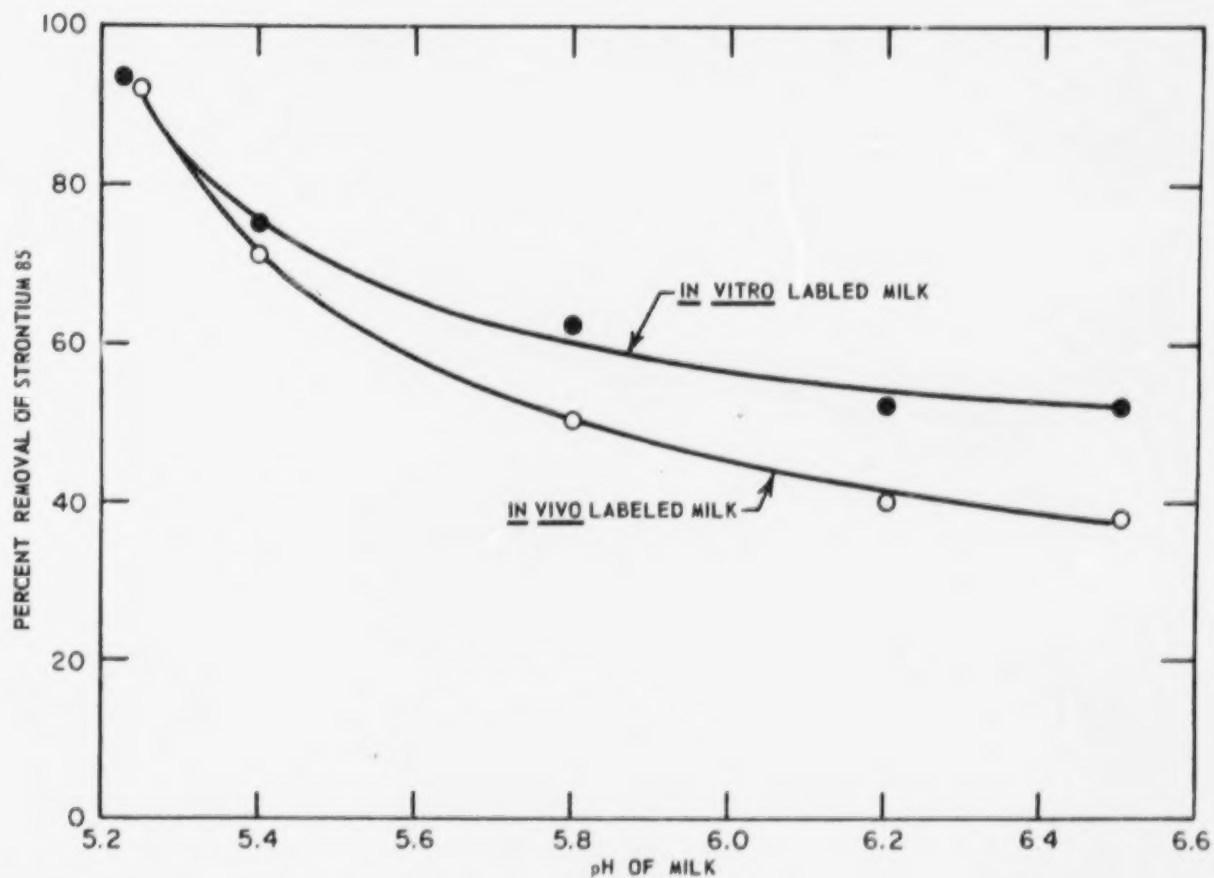


Figure 3. Effect of pH on the removal of ^{85}Sr from milks labeled *in vivo* and *in vitro*

^{85}Sr was removed from both types of labeled milk whereas at higher pH values the amount of strontium 85 removed differed significantly with the labeling method. Therefore, it is reasonable to assume that the results obtained with milk labeled *in vitro*, at pH 5.2 to 5.4, will be essentially the same as those with milk labeled *in vivo*. These limitations of pH apply to removal of protein-bound radionuclides only and not to monovalent cationic radionuclides like cesium.

Effect of number of resin-bed volumes of milk passed through the column on removal of radionuclides

The data presented in figure 4 show a constant removal of 94 percent strontium 85 and 92 percent barium 140 up to 30 resin-bed volumes of milk, beyond which a gradual decrease was observed. The behavior of cesium 134 is quite striking, in that its removal decreased from 98 percent to 0, with increase in number of resin-bed volumes of milk from 5 to 50, revealing that this procedure was unsuitable for the removal of radioactive cesium.

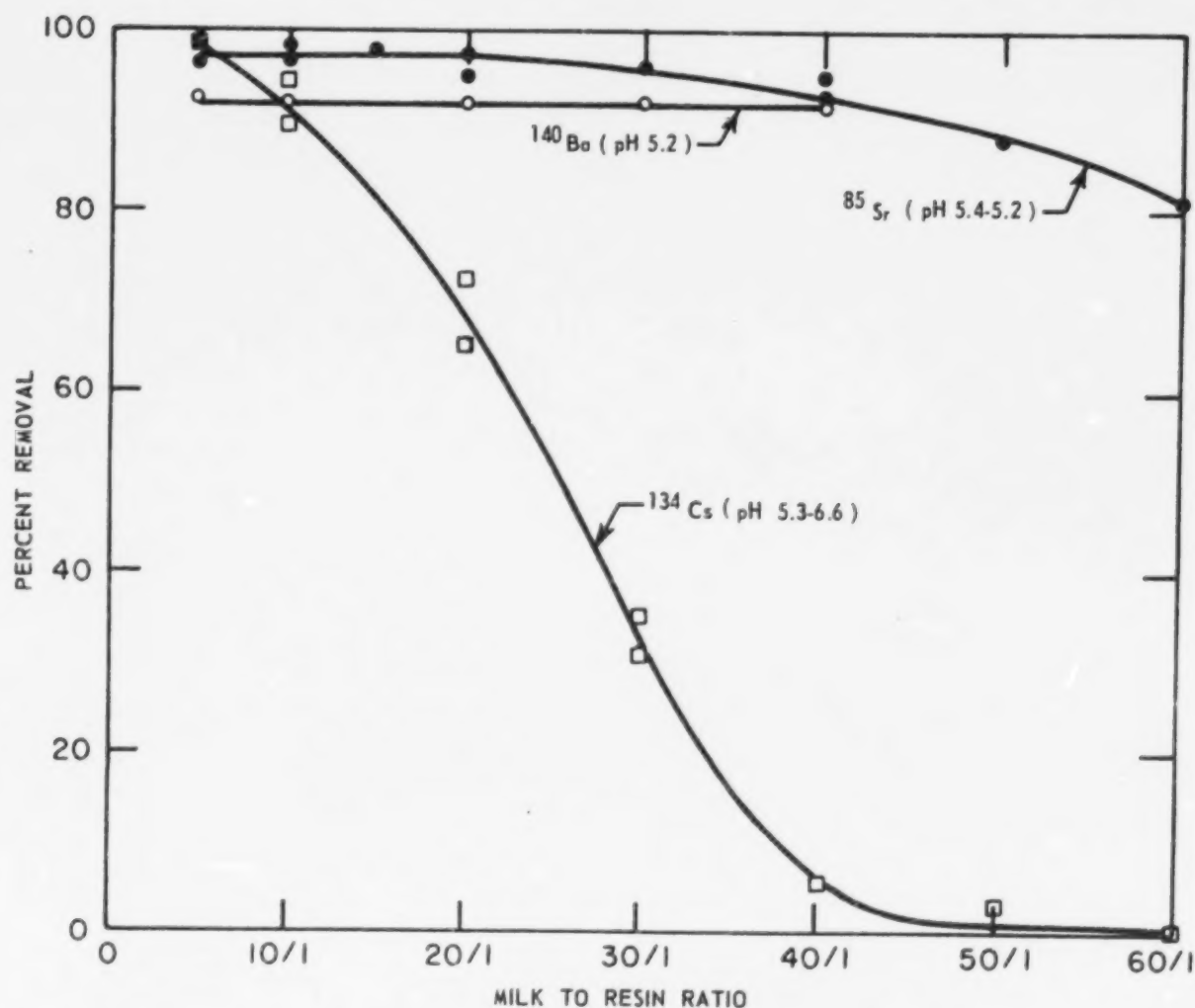


Figure 4. Effect of milk-to-resin ratio on the removal of ^{85}Sr , ^{140}Ba , and ^{134}Cs from milk

Modification of the procedure for the removal of cesium

Since the previously described procedure was not satisfactory for the efficient removal of cesium 134, certain modifications in the process were necessary. The hypothesis was advanced that when citric acid and resin treated milk is neutralized to pH 6.6 with the alkali, the concentration of calcium, magnesium, strontium, and barium in solution is reduced below that in the untreated milk by the complexing effect of these ions with the colloids and the excess citrates. On the other hand, the monovalent cations, such as sodium, potassium, and cesium, do not form any complexes, and therefore would be present as free ions readily available to ion exchange reactions. Figure 5 shows the effect of passing milk, acidified and treated with CaMgKNa resin at 5.4, through a second CaMgKNa column at pH 6.6. From 100 to 80 percent with an average of 90 percent of the residual, cesium 134 was removed.

As the pH of neutralization was increased above 6.6, more cesium 134 was removed (99 percent at pH 7.7), indicating stabilization of the divalent cation complexes with citrates and colloids. However, pH values above 6.6 were considered unsatisfactory from the point of consumer acceptance of the finished product.

This treatment also removed 45 and 30 percent of the residual strontium 85 and barium 140 at pH 6.6, and therefore suggests that neutralization to pH 6.6 does not completely reduce the concentration of divalent cations to be unavailable for ion exchange. The overall effect of these two column treatments resulted in the removal of 95 to 98 percent of strontium 85, 90 to 95 percent of barium 140, and 95 to 100 percent of cesium 134.

Cationic composition of milk

Untreated milk and milk treated with cationic resin were analyzed for the major cations and the results are presented in tables 1 and 2. The data show no significant differences indicating that the method described herein may be successfully employed for the removal of radionuclides from milk.

While these investigations were in progress, the project on radionuclide removal at Beltsville was being established. We had the privilege of training a PHS person who was later to be assigned to Beltsville. The method developed at Cincinnati was transmitted through this person for possible consideration to be adapted to pilot-plant studies.

In fact, the method has proved successful and the USDA people were able to do some organoleptic tests on the milk samples processed according to procedure developed at Cincinnati. The results are presented in table 3. The data show no significant difference in the flavor score between untreated and treated milks, except that the numerical score given is a few points lower than that usually given according to the American Dairy Science Association scoring system.

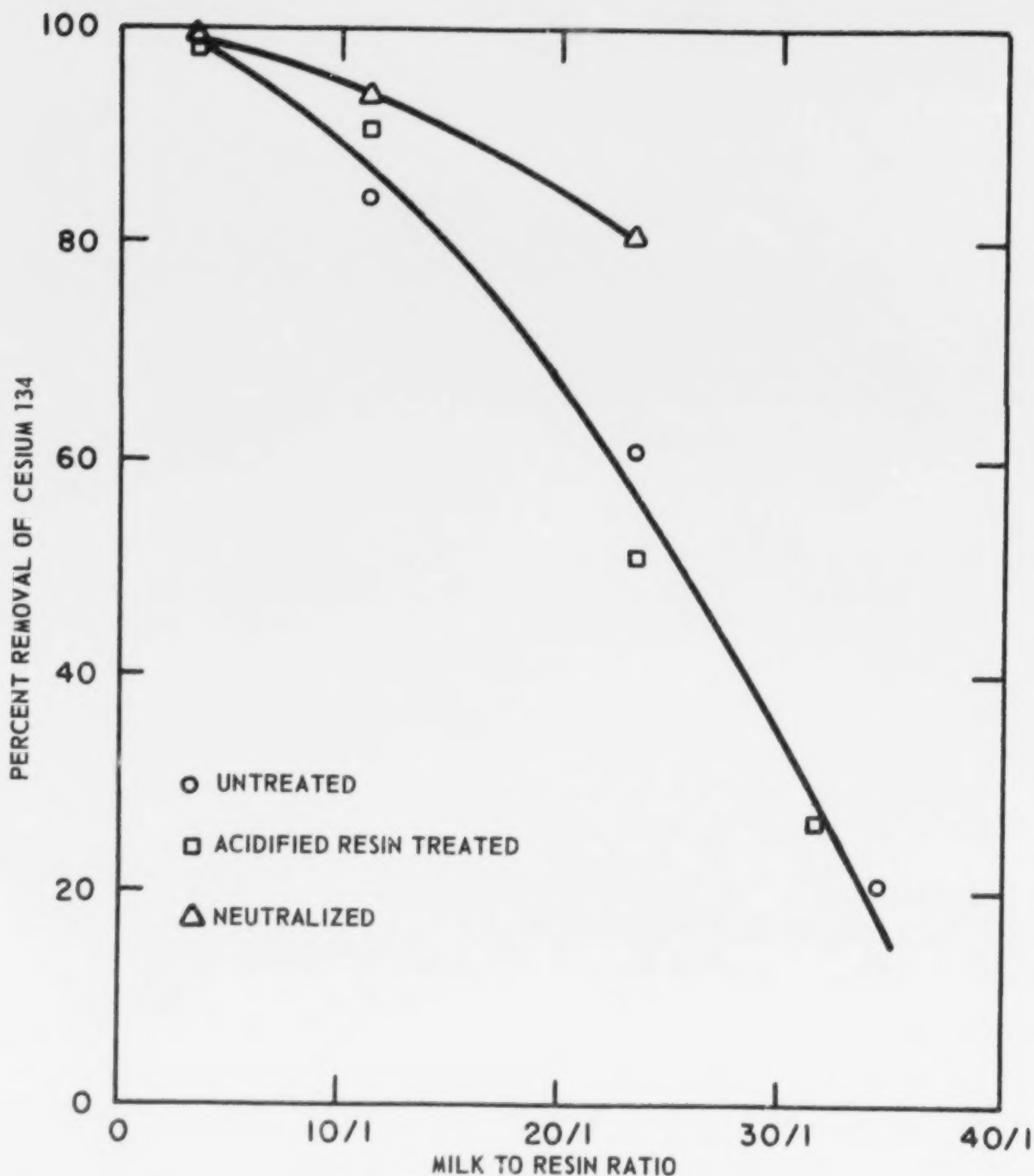


Figure 5. Effect of CaMgKNa resin (pH 5.4) treatment, neutralization to pH 6.6, and CaMgKNa resin (pH 6.6) treatment in succession on ^{134}Cs removal from milk

Table 1. Effect of CaMgKNa resin treatment at pH 5.4 and neutralization to pH 6.6 with OH⁻ resin on the cationic composition of milk

Sample	Treatment	Ca (g/liter)	Mg (g/liter)	K (g/liter)	Na (g/liter)
1-----	Before-----	1. 15	0. 13	1. 36	0. 50
	After-----	1. 14	. 14	1. 38	. 49
2-----	Before-----	1. 12	. 10	1. 32	. 44
	After-----	1. 12	. 10	1. 28	. 44
3-----	Before-----	1. 11	. 19	1. 32	. 49
	After-----	1. 12	. 17	1. 36	. 50
4-----	Before-----	1. 14	. 18	1. 43	. 49
	After-----	1. 14	. 15	1. 42	. 48

Table 2. Effect of CaMgKNa resin treatment at pH 5.4, neutralization to pH 6.6, and CaMgKNa resin treatment at pH 6.6, in succession on the cationic composition of milk

Treatment	Sample	Ca (g/liter)	Mg (g/liter)	K (g/liter)	Na (g/liter)
Column at pH 5.4---	Untreated-----	1. 15	0. 13	1. 36	0. 50
	Treated:				
	1st 100 ml effluent-----	1. 14	. 16	1. 36	. 48
	4th 100 ml effluent-----	1. 13	. 13	1. 38	. 48
	8th 100 ml effluent-----	1. 14	. 14	1. 38	. 50
	12th 100 ml effluent-----	1. 12	. 15	1. 40	. 49
	Composite-----	1. 14	. 14	1. 40	. 49
Column at pH 6.6---	Treated:				
	1st 100 ml effluent-----	1. 15	. 13	1. 32	. 43
	4th 100 ml effluent-----	1. 14	. 15	1. 31	. 43
	8th 100 ml effluent-----	1. 13	. 17	1. 34	. 44
	Composite-----	1. 14	. 15	1. 32	. 43

As Mr. Fooks pointed out earlier, public patent number 3,020,161 has been issued for this process in the names of Drs. Murthy, Masurovsky, Campbell, and Edmondson.

In the next few minutes I would like to briefly outline some of the other work we have been doing in connection with this ion exchange process.

Table 3. Effect of resin treatment on the flavor score of milk

Initial pH	Cation resin treatment	Method of restoring pH	Average flavor score
5.4	No	Anionic resin	35.5
5.4	Yes	Anionic resin	36.0
5.4	Yes	KOH	35.8
5.8	No	Anionic resin	36.0
5.8	Yes	Anionic resin	36.0
5.8	Yes	KOH	35.8
6.2	No	Anionic resin	36.2
6.2	Yes	Anionic resin	35.2
6.2	Yes	KOH	35.6
6.68	No	None	36.6
6.68	Yes	None	36.1

Analysis of milk samples for some of the physical and chemical properties revealed that in comparison to the control milk the treated milk has undergone several treatments like acidification, resin treatment, neutralization, and so forth. Because of this, most of the calcium, including some protein-bound phosphorus, was ionized to be in the soluble form (soluble inorganic and soluble colloidal).

When rennet solution was added to resin-treated milk no coagulation was observed because of the complexing effect of citrates with calcium, thus making calcium unavailable for the formation of coagulum. It was evident that if calcium was added to milk prior to rennet near normal coagulation will occur. Addition of 5 to 6 milliliters of 1M CaCl_2 per liter of milk yielded coagulum comparable to that obtained with untreated milk. This rennet coagulation property was considered very important for cheesemaking.

Several trials were made to prepare cheese from resin-treated milk which indicated that in order to make cheese comparable to that from untreated milk several modifications in the process were necessary: (1) a rennet test must be made to determine the amount of calcium to be added to milk which would yield satisfactory curd; (2) after pasteurization of milk, the milk is brought to 30° C., or if the milk is held in the refrigerator, the milk should be warmed to 40° C., and then cooled to 30° C. prior to addition of culture and incubation in order to avoid fat losses during cheesemaking; and (3) after incubation period, cutting of curd, and prior to cooking the curd, sufficient whey must be removed and replaced with water so as to lower the whey acidity to the same value as that observed with untreated milk whey. The reason for this is that addition of citric acid and potassium hy-

droxide to treated milk increases the titratable acidity by 0.03 to 0.04 percent, and consequently during cooking of cheese curd there is a shrinkage with rapid expulsion of whey and the curd appears tough. Some of these changes in the procedure appear to yield good cheese.

Butter samples have been made from both untreated and resin-treated milks. Figure 6 shows the keeping quality of butter at room temperature. As you can see, the butter from resin-treated milk keeps better than the control sample, possibly explained by the removal of 30 to 40 percent of the copper from milk during resin treatment.

Several cottage cheese samples made from resin-treated milk showed better aroma and taste. This is probably due to the presence of citric acid in the final product. In studying the properties of various products, we find that there is greater binding capacity of casein with the citrates when compared to the control sample.

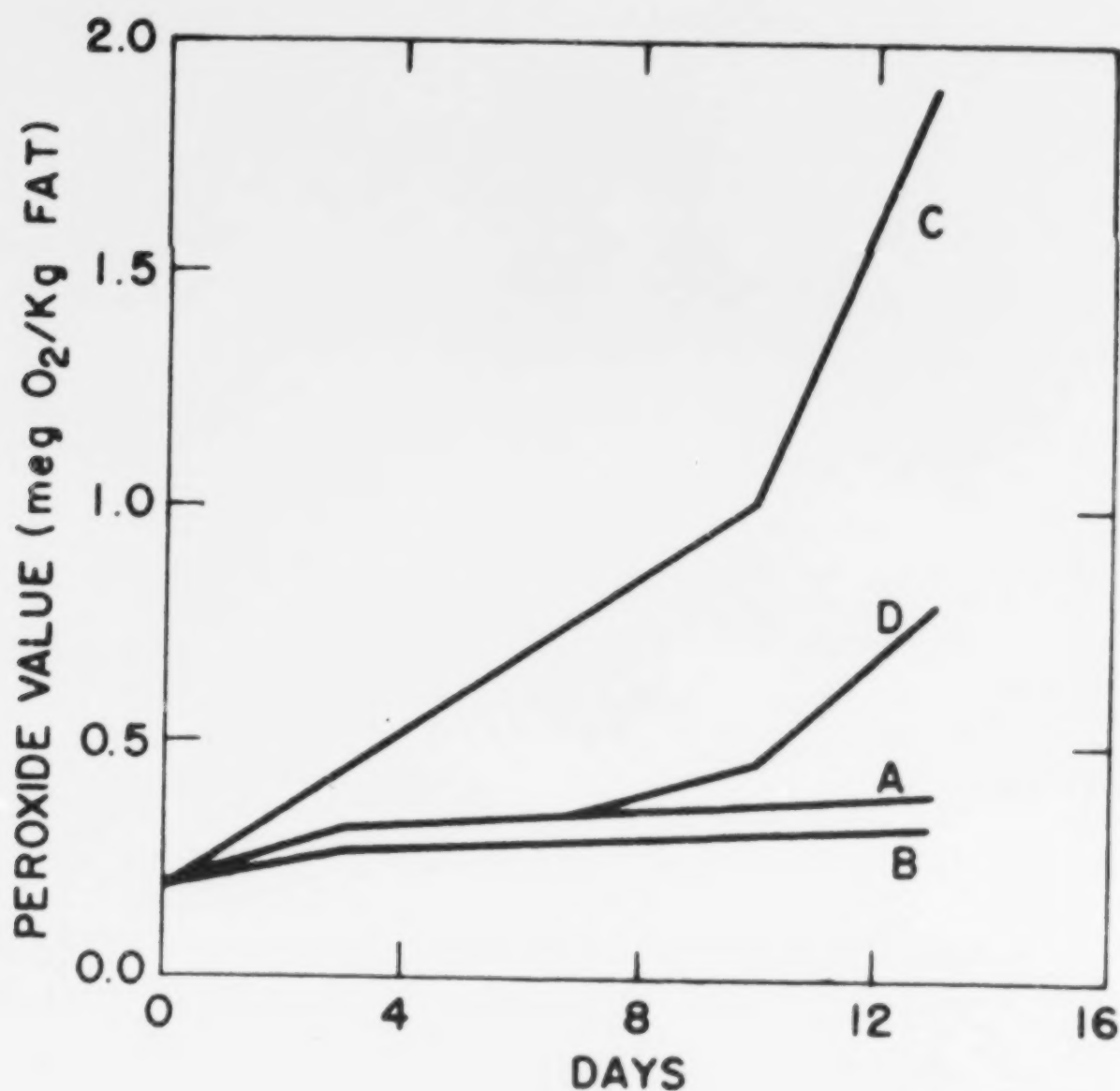


Figure 6. Keeping quality of butter stored at room temperature A = control, unsalted; B = resin treated, unsalted; C = control, salted; and D = resin treated, salted

Attempts were made to prepare cultured buttermilk using commercially available cultures (Meyer-Blanke Co., St. Louis, Mo.). Table 4 shows the titratable acidity and diacetyl content of the samples. The high production of diacetyl in the resin-treated product is due to the presence of citric acid. In addition to high diacetyl content, the samples also showed a high amount of carbon dioxide which was objectionable. By properly selecting the type of culture, it is possible to compare with that made from untreated milk in terms of flavor, titratable acidity, and diacetyl content.

In addition to all the above mentioned investigations, we have also developed a process for the removal of iodine 131 from milk for which we have secured a patent. This process has been tried at the pilot-plant scale level at Beltsville in conjunction with the strontium 90 removal process. We have also done some work on the method whereby neutralization of milk, acidified with citric acid or hydrochloric acid, and treated with CuMgKNa resin to remove iodine 131 and restore the anionic composition of milk in a single process, may be achieved.

Table 4. Diacetyl content of cultured buttermilk made from control and resin-treated milks

Sample	Culture *	Inoculum (percent)	Titratable acidity (percent)	Diacetyl content (ppm)
Control.....	B-1	1	0.76	1.58
	B-1	2	.80	1.82
Treated.....	B-1	1	.77	0.83
	B-1	2	.81	1.86
Control.....	B-2	1	.79	2.83
	B-2	2	.80	2.53
Treated.....	B-2	1	.81	2.18
	B-2	2	.81	2.34
Control.....	B-3	1	.82	2.16
	B-3	2	.86	1.88
Treated.....	B-3	1	.84	8.94
	B-3	2	.87	8.32

Note: Butterfat 3.4 percent, total solids 11.50 percent.

* Source: Meyer-Blanke Co., St. Louis, Mo.

DISCUSSION

Question:

Why wasn't the removal of cesium 137 done prior to acidification?

Dr. Murthy:

The pH has no effect on the removal of cesium 137. The hypothesis was that during neutralization of milk, acidified with citric acid, the calcium citrate complex becomes stabilized and would not be freely available for ion exchange (at higher pH), whereas cesium 137 would be available. We did not try to remove cesium 137 first and then acidify milk for removing strontium 90.

Mr. Terrill:

I think Dr. Edmondson might have some comments on that because in the next phase of the pilot-plant operation these things were perhaps considered more in detail.

Are there any other questions or comments from anyone?

Thank you, Dr. Murthy. I know everyone appreciates not only the comments here but the excellent work done by you and others at Cincinnati.

THE BELTSVILLE PROCESS

*L. F. Edmondson*¹

My presentation reviews the research and development program which was carried on at Beltsville for about 4 years. I will describe primarily our fixed-bed, ion exchange procedure for removing strontium 90 from milk, and discuss some of the factors affecting the removal of radionuclides, means of controlling the composition of milk, and other procedures for removing radionuclides. Our objective in this work was "to develop a feasible process for removing radioactive contamination from milk." We think the developments through the pilot-plant stage are satisfactory; consequently, this phase of the joint project was terminated this past summer.

Figure 1 shows schematically the pilot-plant equipment used for removing radiostrontium from milk. This might be considered a prototype of the commercial scale process which you will see here. Some of the important equipment units are not indicated, particularly the control panel, milk filters and "in-line" pH electrodes. However, the flow sheet is sufficient to follow the characteristic steps in the process. A similar scheme was published a few years ago (1); however, most of the present pilot-plant equipment is new, including a large milk-holding tank, stainless steel columns, continuous in-line acidification and neutralization equipment, automatically controlled valves which are arranged so that the milk, rinse water, detergent solution, and mixed salt regenerating solutions can be passed through the resin bed in the proper sequence. Four 200-gallon regenerant tanks provide for the reuse of three-fourths of the salt solution (only three tanks are shown in fig. 1). The pilot plant was designed for a capacity of 100 gallons per hour.

The process includes the following operations:

1. Initial resin preparation. The resin is regenerated initially with a mixed salt solution of calcium, magnesium, potassium, and sodium

¹ Eastern Utilization Research and Development Division, Agricultural Research Service, U.S. Department of Agriculture.

chlorides. The composition of the solution used during most of the pilot-plant development work was (in grams per liter): $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, 53.5; KCl , 23.1; NaCl , 8.5; and $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, 15.1. These quantities were selected so that the resin would be equilibrated with respect to the major milk cations, thus assuring a minimum of change in the mineral composition of the resin-treated milk. The factors affecting the optimum composition, and a procedure for selecting the relative proportion of each salt are considered in two previously published papers (2-3). About 10 resin-bed volumes of the fresh salt solution should be sufficient for the initial regeneration, the exact quantity depending on the temperature and flow rate of the regenerant.

2. Rinse excess salt solution from column thoroughly with water.

3. Acidification of milk. The cold, raw, whole milk is adjusted to 50°F ., then acidified to pH 5.3 by an acid-metering pump with a solution of $0.75M$ citric acid (fig. 1). The increased removal obtained by acidification and explanations for the effect are discussed in a number of papers (4-7).

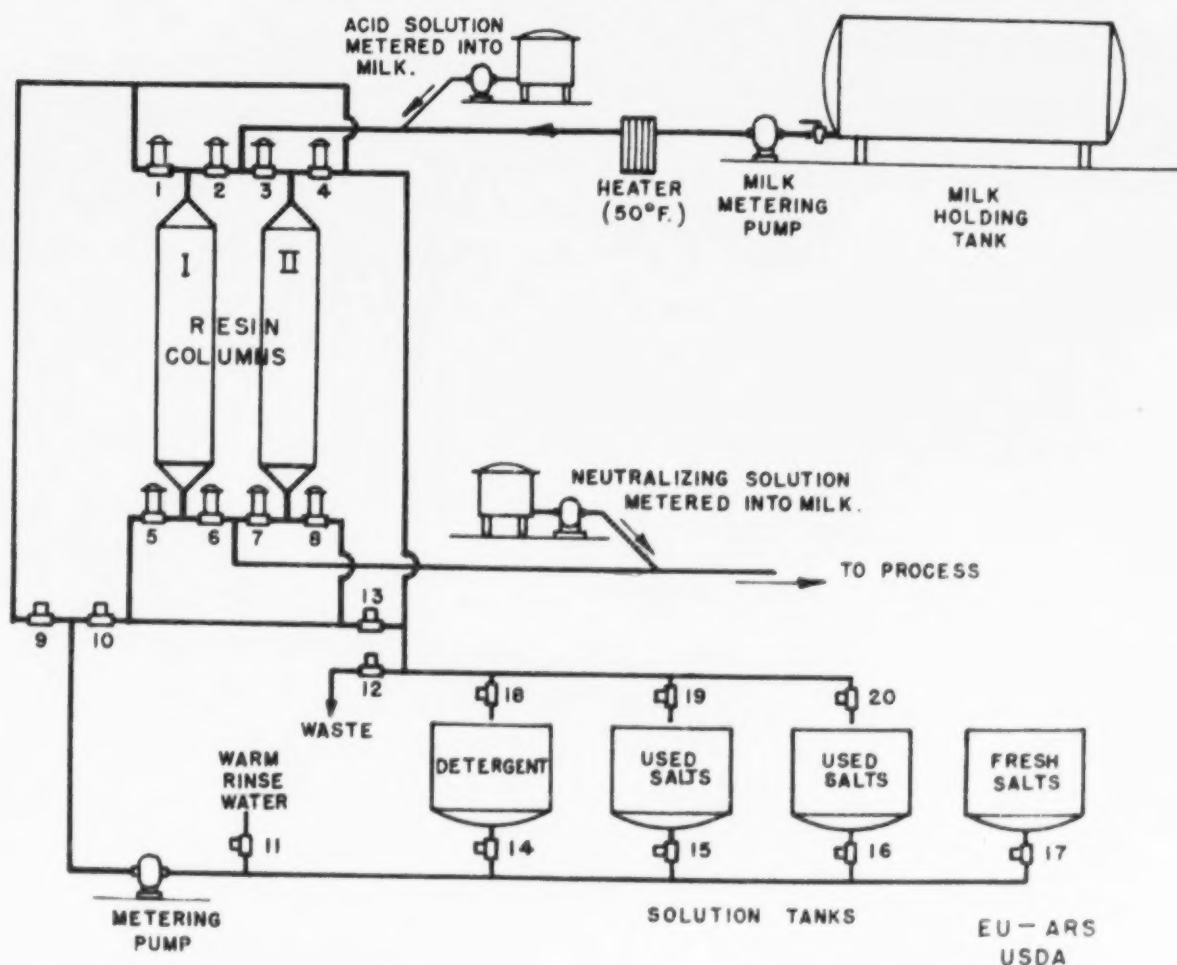


Figure 1. Beltsville ion exchange process for removal of radiostrontium from milk

Citric acid has been found to effect a greater removal of radiostrontium than either HCl or H_3PO_4 (5). The milk is then filtered to remove any fine material which might cause plugging in the resin column.

4. Flow of milk through the resin column. Referring to figure 1, the milk would flow through Column I by opening valves 2 and 6. After a period of time (up to 8 hours of continuous flow) the milk may be diverted to Column II by closing valves 2 and 6, and opening valves 3 and 7. Column I may then be rinsed, sanitized, and regenerated. The effect of flow rate of milk through the resin bed on the removal of radiostrontium is discussed in another paper (4). For practical use, either of two flow rates are suggested: $\frac{1}{8}$ and $\frac{1}{16}$ resin-bed volumes (rbv) per minute. For a capacity of 100 gallons per hour, these rates require backwashed and settled resin volumes in the column of 13.35 and 26.7 gallons, respectively. The resin inventory per 8 hours of processing time, and regenerant requirements per unit volume of milk will be the same in either case. Thirty resin-bed volumes of milk flowing through a column require 8 hours at $\frac{1}{16}$ rbv per min. and 4 hours at $\frac{1}{8}$ rbv per min.

5. Neutralization and processing of the treated milk. The effluent milk from the columns is neutralized by continuous inline addition of a solution of 1.5N KOH, or with other bases. It may then be pasteurized and homogenized in the usual manner.

6. Cleaning and sanitizing the resin. After milk is diverted to Column II (fig. 1), Column I may be rinsed by opening valves 11, 9, 1, 5, 13, and 12. To wash upflow through the column with a detergent solution, valves 11 and 9 are closed; valves 14, 10, 5, 1, and 18 are opened. If the detergent is directed to "Waste," valve 12 instead of 18 is opened. After another water rinse the column is ready for regeneration.

7. Regeneration of the resin is carried out with the same salt solution used for the initial preparation. Its chief purpose is to remove the radiostrontium from the resin; it also maintains equilibrium with respect to the minerals in milk. It is economically desirable to reuse as much of the regenerant as possible. The Beltsville process provides for reusing three-fourths of this solution. The first fraction (one-fourth of the total) through the column is discarded. Each of the three remaining fractions is pumped into separate tanks, which are used for subsequent regeneration in the sequence in which they are recovered. The fourth tank in the series is filled with fresh solution equal in volume to the amount discarded from the first tank in the series.

Laboratory data have shown that, under steady-state conditions, each fraction "strips" approximately the following percentage of radiostrontium from a column:

Fraction number	Percent of total activity
1-----	65.4
2-----	25.9
3-----	6.7
4-----	2.0

The total amount in the first fraction, of course, will be equal to 100 percent of the quantity removed, including accumulations during previous use. Obviously, it would be wasteful to discard tanks 2, 3, and 4.

The regenerant is passed downflow through the column by opening the appropriate valves indicated in figure 1. For the process described above, about 16 rbv of regenerant are required, 4 rbv of which are fresh solution. Thus, 1 volume of salt solution is required for each 7.5 volumes of milk. The effectiveness of the regenerant for removing radionuclides from a resin is increased by heating (8). A further advantage of heating this solution is in the reduction of bacterial growth in the resin columns. The advantages gained may be partially offset by the expense of heating.

The following table outlines the steps and suggested time schedules for an 8-hour milk cycle through one column without interruption.

Step	Quantity (rbv)*	Time (min)	Temperature (F.)
Rinse water-----	6	24	50
Milk-----	30	480	50
Rinse water-----	6	24	100
Detergent-----	8	32	150
Rinse-----	6	24	160
Regenerant No. 1-----	4	40	70
Regenerant No. 2-----	4	40	70
Regenerant No. 3-----	4	40	70
Fresh regenerant-----	4	40	70
Rinse water-----	6	24	160

* One resin-bed volume equals 26.7 gallons.

The above schedule is subject to change, depending on additional studies being carried out by the Public Health Service on sanitation procedures and on the experience gained during commercial test runs.

PILOT-PLANT TEST RUNS FOR REMOVING STRONTIUM 90 FROM MILK CONTAINING ONLY ENVIRONMENTAL LEVELS

Practically all of the research carried out during the development of the fixed-bed cation process was done by using strontium 85 because the method for its assay is simple and rapid compared to that for strontium 90. Levels of about 1 microcurie per quart of milk were normally used for the removal studies. This compares with about 25 picocuries of strontium 90 in current milk sources; that is, the levels of strontium 85 were about 40,000 times the environmental levels of strontium 90. The percentage removal of strontium 85 from milk labeled *in vivo* and of strontium 90 should be the same. This was confirmed by determining the percentage of strontium 90 removed from milk obtained from a local dairy. The average removal from three separate runs of 30 resin-bed volumes each, processed in the pilot plant, was 95 percent. This is in good agreement with data obtained in the laboratory with the high levels of strontium 85.

Only small changes in the flavor of the resin-treated milk have been observed. Flavor scores, as judged by a panel of 10 judges, have usually averaged about 1.0 point below the controls on the American Dairy Science Association scorecard.

COST OF THE PROCESS

The cost of operating the fixed-bed process as described above is estimated at 1.0 cents per quart of milk when USP grades of citric acid, potassium hydroxide, and chloride salts of calcium, potassium, sodium, and magnesium are used. If technical grades of the chloride salts are used for regeneration, the cost is estimated at 0.5 cents per quart. These costs are based on pilot-plant experience and include estimates for investment, investment burden, labor and normal operating costs.

FIXED-BED PROCESS FOR REMOVING BOTH IODINE 131 AND RADIOSTRONTIUM

A strong base, anion exchange resin may be used to remove iodine 131 under conditions similar to those used for the removal of cationic radionuclides. The resin is regenerated with a mixed solution of the sodium salts of the major milk anions—NaCl, Na₂HPO₄, and sodium citrate (9). An integrated pilot plant process was arranged for removing both iodine 131 and the cationic radionuclides by passing milk first through the anion resin (Dowex 2×8) which was contained in 6-inch glass columns, then acidifying to pH 5.3, and continuing the milk flow through the cationic resin column. Over 90 percent of the iodine was removed with retention of an excellent flavor.

INVESTIGATIONS WITH CONTINUOUS ION EXCHANGE PROCESSES

Another phase of the isotope removal investigations included the use of a continuous ion exchange contactor. It was a laboratory model having a capacity of about 12 gallons per hour. This system was used to study (1) the removal of cationic radionuclides (strontium 85 and cesium 134) from milk using a strong acid resin in the mixed form, (2) the removal of strontium 85 with the resin in the sodium form, and (3) the removal of iodine 131 and simultaneous neutralization with a strong base resin in the hydroxyl form. The latter procedure was studied with the intention of using it as a second stage in a cation-anion removal process. Additional discussion of the continuous processes may be found in a previous publication (10).

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NUTRITIONAL INVESTIGATION OF PILOT-PLANT PROCESSED MILK

Presented by DeWitt G. Hazzard^{1, 2}

INTRODUCTION

Mr. Terrill:

I think we should call to your attention that in areas such as nutrition our investigations have to be largely related to quality per unit of volume or per unit of consumption. The question would be how much the introduction of a process like this would influence the total intake of milk and milk products, whether it would go up or down, and under what circumstances. I think you folks are the best judges of this. If we take an extreme case, such as very intense atomic fallout or atomic war, I think most of us would quickly agree that a process such as this to remove strontium 90 from the milk would be, in fact, helpful in retaining milk as one of our basic nutrients. On the other

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hand, I think, if we should introduce this process too early in the game, it might result in a decrease in the consumption of milk and possibly lower the nutritional benefits which we get from milk, even though per unit of milk consumed would be highly satisfactory. I think the question is, at what point should a change be considered. In this area, many of you representing the industry and the public health and agricultural agencies are in a better position really than we, who are concerned with developing the process *per se*. I think the same thing tends to apply to sanitary aspects, and even though we are developing data on economics that can tell us within reason what the cost is per unit volume, the overall economic effects of the introduction of this process under several conditions will again largely be a matter of judgment. I would urge you to keep these things in mind while these next discussions go forward.

At this time I would like to call on Dr. Hazzard, Division of Radiological Health, who has been responsible for some of the nutritional investigations.

Dr. Hazzard:

I think by this time you all have a pretty good idea of what this process is all about and I need not go into much background material, but get straight to the point of my topic, which is the nutritional investigation of the quality of milk following treatment. The assessment of nutritional quality of milk following the treatment has been attempted by two approaches: (1) a direct chemical analysis of milk, and (2) feeding studies and subsequent biochemical analyses. The control milk for this study was prepared by standardizing the raw whole milk to 3.5 percent butterfat, pasteurizing at 161° to 165° F. for 15 seconds, and homogenizing at 2,000 pounds per square inch. The resin-treated milk was likewise standardized to 3.5 percent butterfat and then subjected to the cation exchange process. Samples of both control and processed fluid whole milk were taken at this point for chemical analyses, and the remainder of the milk was spray dried and packaged for later feeding studies.

Let us first look at the direct chemical analyses assessment. These were initiated in November 1961 under the direction of Dr. Russell Isaacks, formerly of the Division of Radiological Health. As can be seen in table 1, these data indicated no significant differences at the 5-percent level in total solids, butterfat, crude protein, or flavor score, but there was an increase of 12 percent in the ash. This was significant at the 5-percent level. The concentrations of the four major cations

were unchanged in the resin process, but the subsequent KOH neutralization resulted in an approximate 80-percent increase in the potassium content of the consumable product (table 2).

Table 1. Gross chemical analyses

Determination	Milk	
	Control	Processed
Total solids (percent) -----	12. 79	12. 41
Butterfat (percent) -----	3. 63	3. 50
Crude protein (percent) -----	3. 13	3. 15
Ash (percent) -----	. 70	. 80
Flavor score -----	36. 6	35. 8

Table 2. Mineral analyses

Element	Control (mg/liter)	Processed (mg/liter)
Ca -----	1, 242. 00	1, 187. 00
Mg -----	107. 00	110. 00
Na -----	479. 00	491. 00
K -----	1, 391. 00	2, 503. 00
P -----	855. 00	874. 00
Fe -----	. 75	. 74
Cu -----	. 23	. 16

Decreased copper and increased phosphorous concentrations of the process milk were shown (these were not significant at the 5-percent levels), while the iron content remained unaltered. Although it was not significant, there was a decrease of 0.1 percent in the butterfat content of the treated milk. A check was therefore made of the fatty acid distribution of the milk (table 3). This, likewise, showed no appreciable changes. The carotene, vitamin A, riboflavin, pantothenic acid, folic acid, and vitamin B₁₂ concentrations in the process milk were not significantly different from those of the control milk (table 4). The concentrations of thiamine, niacin, and vitamin B₆ were all significantly reduced in the processed milk by 50, 27, and 15 percent, respectively. It was rather interesting that only 15-percent removal of the vitamin B₆ occurred since the ion exchange resin used in this process is the same one used for the chromatographic separation of the

three forms of vitamin B₆. However, this can be explained by the fact that in such a separation the sample is previously autoclaved, thereby liberating the vitamin from its organic bonds, whereas in the process the vitamin is organically bound.

Table 3. Fatty acid composition

Fatty acid ^a	Control (percent)	Processed (percent)
10:0-----	^b T	^b T
12:0-----	3.2	1.9
14:0-----	15.7	14.3
15:0-----	1.6	1.5
16:0-----	42.1	44.0
16:1-----	1.1	1.1
18:0-----	9.5	10.2
18:1-----	24.8	25.0
18:2-----	1.4	1.4
18:3-----	1.0	1.0

^a Number preceding colon is carbon chain length; number following colon is number of double bonds.

^b T indicates trace.

Table 4. Vitamin analyses

Compound	Control (μg/liter)	Processed (μg/liter)
Carotene-----	164.50	157.5
Vitamin A-----	47.3	45.5
Thiamine (total)-----	275.0	140.4
free thiamine-----	137.5	19.3
bound thiamine-----	137.5	121.1
Riboflavin-----	1,250.0	1,340.0
Pantothenic acid-----	2,580.0	2,670.0
Niacin-----	840.0	610.0
Folic acid-----	9.4	8.6
Vitamin B ₁₂ -----	4.6	4.4
Vitamin B ₆ (total)-----	390.0	330.0
pyridoxine-----	20.0	10.0
pyridoxal-----	290.0	250.0
pyridoxamine-----	80.0	70.0

The data on thiamine again indicate that the organically bound forms are unavailable for absorption. After noting the approximate 50-percent reduction in thiamine in milk exposed to the cation exchange by this and other groups, it was decided to investigate more closely the two forms of thiamine. It can be seen from figure 1 that the free and bound forms of thiamine are about equally divided in the control milk; however, after two to six bed volumes have passed through the resin 61 percent of the total, composed of 93 percent free and 37 percent bound, was removed. Corresponding samples collected from the treated milk between the 10th to 14th bed volumes contained 47 percent of the total, and were composed of 85 percent of the free and 19 percent of the bound forms. The fraction between the 24th and 48th bed volumes contained 41 percent of the total, composed of 83 percent free and 8 percent bound forms. Approximately 69 percent of the thiamine removed was recovered by elution from the resin. It is possible that even greater recovery could be achieved with different solvents and greater volumes. However, this serves to indicate that at least a considerable amount of the thiamine is absorbed on the resin and not actually destroyed.

Let us now look at the second approach to the nutritional assessment, that of the feeding studies. It was thought that assessment of alterations by feeding studies would indicate not only the severity of

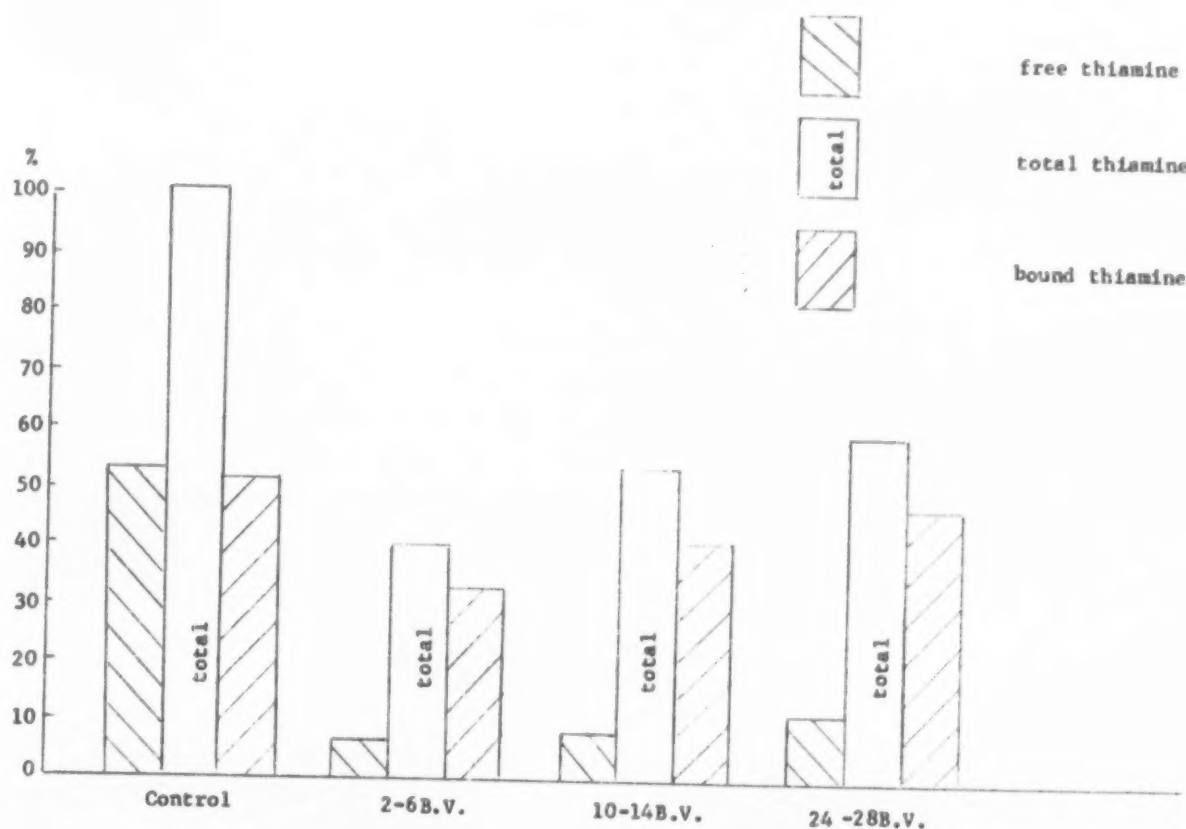


Figure 1. Effect of length of run on thiamine removal

the observed chemical alterations, but would serve as an indicator of alterations in composition which were not determined through the chemical analysis. All milk in these studies was the spray-dried powder previously described. These studies (table 5) consisted of three separate experiments, one using month-old rats, and two using baby pigs. The reason for the use of baby pigs is that supposedly their digestive system at this age is very similar to that of the human. In the rat study, six male and six female albino litter mates were randomly assigned to each of six groups and replicated seven times for a total of 84 rats. Rats were maintained in individual cages with screened floors and received food and water *ad libitum*. Although milk has often been termed the universal food because of its nutritious qualities, it has been known for years that bovine fluid milk alone would not support normal growth in rats. Previous research has shown that such milk must be supplemented by copper, iron, manganese, and for rats over 7 to 8 weeks of age, with a supplemental energy source in order to maintain normal growth. Based upon these earlier findings, the diets shown in table 6 were used. The vitamin levels used in diet I were approximately $\frac{1}{10}$ to $\frac{1}{15}$ normal, while those of diet II were double the quantities in diet I. The mineral levels of diet I were $\frac{1}{4}$ normal while those of diet II were again doubled. The purpose of such regimens was to stress the animals with suboptimal, marginal, and optimal levels of both vitamins and minerals.

**Table 5. Experimental design
(weanling rat study)**

Group	Treatment
1-----	Control milk + (Fe, Cu, Mn)*
2-----	Processed milk + (Fe, Cu, Mn)*
3-----	Diets I + control milk
4-----	Diets I + processed milk
5-----	Diets II + control milk
6-----	Diets II + processed milk

* Trace mineral mixture supplement supplied per kilogram of dried, whole-milk powder: 40 mg copper (157.06 mg $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$), 80 mg iron (398.34 mg $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), 40 mg manganese (162.39 mg $\text{MnSO}_4 \cdot 4\text{H}_2\text{O}$).

In baby pig experiment I (table 7) 3-day-old crossbred piglets were housed in raised screen-floor pens under heat lamps. Litter mates of each sex were distributed at random among three groups, each consisting of two lots of six pigs each, for a total of 36 pigs. A mineral supplement containing iron, copper, manganese, and zinc was added to

all three diets. The milk was reconstituted twice daily and each lot of six piglets was fed on the basis of grams of milk powder per piglet per feeding from a nipples self-feeder. This study lasted 8 weeks, and during the eighth week the piglets were placed in large rabbit metabolism cages for 24-hour urine collections.

**Table 6. Experimental diets
(weanling rat study)**

Ingredients	Diet I (percent)	Diet II (percent)
Casein (vitamin free)-----	13.10	13.10
Dried, whole milk (either control or processed)-----	50.00	50.00
Cerelose (glucose monohydrate)-----	31.55	29.30
Wood pulp-----	3.00	3.00
Choline chloride-----	0.10	0.10
Vitamin mixture ^a -----	1.00	2.00
Mineral mixture ^b -----	1.25	2.50
Total-----	100.00	100.00

^a Vitamin mixture supplement supplied per kilogram of diet: 1.0 mg thiamine hydrochloride, 1.5 mg riboflavin, 1.5 mg pyridoxine hydrochloride, 10.0 mg calcium pantothenate, 0.2 mg folic acid, 20.0 mg nicotinic acid, 20.0 mg inositol, 120.0 mg p-aminobenzoic acid, 0.05 mg vitamin B₁₂, 7.50 mg menadione 0.10 mg biotin, 2,000.0 mg DL-methionine, 2.0 mg alpha tocopheryl acetate, 30,000 I.U. vitamin A, and 4,000 I.U. vitamin D in 7.816 grams of cerelose as carrier.

^b Mineral mixture supplement supplied per kilogram of diet: 2.67 grams CaCO₃, 4.16 grams CaHPO₄·2H₂O, 0.25 gram MgCO₃, 130 grams MgSO₄·7H₂O, 1.27 grams NaCl, 0.21 gram KCl, 3.15 grams KH₂PO₄, 0.20 gram FeSO₄·7H₂O, 0.25 gram MnSO₄·4H₂O, 0.006 gram KI, 0.016 gram CuSO₄·5H₂O, 0.010 gram NaF, 0.0024 gram K₂Al₂(SO₄)₄·24H₂O, 0.005 gram ZnSO₄·7H₂O, and 0.0006 gram CoCl₂·6H₂O.

**Table 7. Experimental design
(baby pig studies)**

Group	Treatment
1-----	Control milk powder + (Fe, Cu, Mn, Zn) ^a
2-----	Processed milk powder + (Fe, Cu, Mn, Zn) ^a
3-----	Control milk powder + (Fe, Cu, Mn, Zn) + potassium citrate ^b

^a Trace mineral mixture supplement supplied per kilogram of milk powder: 117.9 mg iron (0.5896 gram FeSO₄), 3.1 mg copper (0.0121 gram CuSO₄·5H₂O), 21.7 mg manganese (0.0598 gram Mn₄·4H₂O), 50.5 mg zinc (0.0629 gram ZnO), and 20.0 mg oxytetracycline.

^b Potassium citrate was added at the level of 29.38 grams per kilogram of milk powder which was equivalent to the level of potassium and citric acid added during the process.

In baby pig experiment II, the animals used were similar to those of the first study. Again, three groups, each with two lots were used; however, each lot consisted of four piglets instead of the previous six, and each piglet was individually housed instead of being placed in groups of six as before. This procedure was used because of the extreme competition, and, I say, extreme for food which was observed in the first study among the six pigs in each lot. Also, instead of starting the experiment immediately at 3 days of age, all pigs were maintained on control milk for 1 week and started on experiment at 10 days of age. This was to allow us time to get the pigs on full feed since in the first study we observed a little trouble in getting them started nursing. This study lasted 4 weeks, with a urine collection during the final week. In both studies, blood samples were taken by *vena cava* puncture.

The third treatment (that of potassium citrate, at a level calculated to be equivalent to the potassium and citric acid added during the resin treatment process) was used to test the metabolic and renal capacity of the baby pig for this level of potassium and citrate, uncomplicated by any known alteration in the whole milk.

Let us take a look at the results of these feeding studies. Growth rates (table 8) of rats showed no significant difference between group comparisons, group 1 versus 2, 3 versus 4, 5 versus 6, for either males or females. There were no significant differences in feed consumption (table 9); however, an unexplainable difference did exist between groups 5 and 6 with regard to water consumption. This was significant at the 5-percent level, and I see no reason for it at all. Maybe this is the five out of a hundred chance; that's about the best explanation I can give for it.

**Table 8. Average body weights of rats (male and female)
(grams)**

Group	Weeks			
	0	4	8	12
1-----	56	173	242	270
2-----	61	178	234	270
3-----	56	190	252	292
4-----	56	192	263	302
5-----	57	191	254	287
6-----	57	184	238	281

Table 9. Average food and water consumption of individual rats (grams)

Group	Food	Water
1-----	894	2,394
2-----	867	2,236
3-----	1,037	2,130
4-----	1,088	2,076
5-----	1,093	2,642
6-----	1,106	2,365

There were no significant differences between treatment in concentrations of sodium, potassium, or calcium in either the blood (table 10) or urine (table 11) of the rats.

Table 10. Rat serum analyses (12 weeks)

Group	Na (mg/100 ml)	K (mg/100 ml)	Ca (mg/100 ml)
1-----	384	13.2	16.3
2-----	391	13.0	15.8
3-----	385	13.7	17.2
4-----	386	12.4	17.4
5-----	396	11.2	18.0
6-----	391	11.8	17.8

Table 11. Rat urine analyses (12 weeks)

Group	Na (mg/24 hr)	K (mg/24 hr)	Ca (mg/24 hr)
1-----	2.8	286	0.19
2-----	3.6	465	.24
3-----	3.2	246	.18
4-----	2.2	334	.20
5-----	4.5	270	.33
6-----	3.4	390	.22

The growth data from both pig studies (table 12) showed no significant differences at either 4 or 8 weeks between any treatments. There were no significant differences in the blood serum concentrations (table 13) of sodium, potassium, or calcium between the three experimental groups. In pig experiment I, there was a significantly greater excretion of both potassium and sodium in the urine at the 5-percent level (table 14) from piglets fed the added potassium citrate. In the second study there was again a significantly greater excretion of urine potassium at the 1-percent level, but no difference in sodium. Pathological observations on four pigs sacrificed from each of the three groups in experiment II at 5 weeks of age indicated no abnormal microscopic findings in the kidneys, livers, or hearts of these animals.

Table 12. Weight gain of pigs

Group		4 weeks (kg)	8 weeks (kg)
1	Experiment 1.....	3.30	13.04
	Experiment 2.....	2.31	—
2	Experiment 1.....	2.59	12.28
	Experiment 2.....	2.83	—
3	Experiment 1.....	2.81	11.97
	Experiment 2.....	2.40	—

**Table 13. Pig serum analyses
(mg/100 ml)**

Group	Na		K		Ca		Mg	
	4 weeks	8 weeks	4 weeks	8 weeks	4 weeks	8 weeks	4 weeks	8 weeks
1 A ^a B ^b	348	400	26.0	12.5	10.7	13.7	1.74	1.80
2 A..... B.....	343	379	26.8	12.4	10.6	13.2	1.49	2.10
3 A..... B.....	347	394	25.3	12.4	10.8	13.1	.57	2.00

^a Experiment No. 1.

^b Experiment No. 2.

Table 14. Pig urine analyses
(mg/24 hr)

Group	Na		K		Ca	
	4 weeks	8 weeks	4 weeks	8 weeks	4 weeks	8 weeks
1 A ^a -----	157	261	952	2,930	—	19.9
B ^b -----						
2 A -----	198	415	1,610	3,558	—	19.0
B -----						
3 A -----	202	776	2,268	5,550	—	28.1
B -----						

^a Experiment No. 1.

^b Experiment No. 2.

Therefore, it would appear that the 80-percent increase in potassium content had no deleterious effects on the animals and that they were capable, through renal excretion, of maintaining normal electrolyte balance in the blood. The reduction of thiamine, niacin, and vitamin B₆ in the processed milk showed no deleterious effects in any of the parameters measured. Calculations indicated that thiamine and vitamin B₆ were present at marginal National Research Council recommended levels, while niacin levels were considerably below requirement. It should be noted, however, that niacin levels of the control milk were likewise deficient, according to National Research Council recommendations.

In summary, chemical analyses of the milk showed the only major changes to be increased potassium concentration and ash content and reduced thiamine, niacin, and B₆ concentration. Animal feeding studies involving 84 rats and 60 baby pigs indicated no deleterious effects on growth or general health as a result of known changes in the processed milk. These findings have all been based on the cation work. I've noted the others have been making some comment on some of the anion work. I might mention here that we have run a very brief chemical analysis on some of the anion milk, analyzing sodium, potassium, calcium, magnesium, and thiamine, and we found no differences between control and processed milk in these five constituents.

DISCUSSION**Mr. Terrill:**

Are there any questions on this paper, or any comments or suggestions?

Question:

Do pigs manufacture niacine as do rats?

Answer:

Right off the top of my head, I don't think I can give you a reliable answer to that! I am not sure without checking.

Mr. Terrill:

I think we should mention at this time, just so it is mentioned, that we have taken up the matter of the use of the ion exchange resin with the Food and Drug Administration, and they have approved it for emergency use. But we have an obligation to keep them fully informed of these investigations, and depending on their judgment, anything we do in this area that tends to add or take away food values will have to be cleared with them, among others

MICROBIOLOGICAL ASPECTS OF THE FIXED-BED PROCESS FOR THE REMOVAL OF CATIONIC RADIONUCLIDES

R. B. Read, Jr.¹

INTRODUCTION

Mr. Terrill:

At this time, we are going to have some discussion of the sanitary aspects of the process. Practically everyone in this room recognizes that when you are doing something that increases the surface contamination of milk, you are working in contradiction to the kinds of procedures you always try to establish in milk plants. We have to be very careful about these things, not only in terms of the planning, but in terms of operations, because it may well be that the things we do here under comparatively well-controlled conditions will not work out quite so well in some of the plants. On the other hand, they might become so standardized that they would work out better than we predict, but this is up to you to judge. At this time, I would like to call on Dr. Read who has been undertaking the sanitary investigations of the process.

Dr. Read:

Once the chemical aspects of the cationic removal process had been examined in some depth, both at the laboratory and pilot-plant levels, studies were started on the microbiology of this process. Two 6-inch-diameter glass columns were set up in the Milk and Food Research pilot plant at the Robert A. Taft Sanitary Engineering Center in Cincinnati for this study. The equipment and the initial operating

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procedures essentially duplicated those developed by the U.S. Department of Agriculture in their study at Beltsville, Md. The process involved batch acidification of milk at 50° F. with citric acid to pH 5.35, pumping the milk through a filter and the resin column with a positive displacement pump, and cooling the milk to 40° F. after it left the resin column. The milk was then brought back to its original pH with potassium hydroxide, using a batch technique. The pumping speeds were set so that 200 gallons of milk could be processed in one 6-inch column, with a resin depth of 54 inches, in 2, 4, or 8 hours for respective pumping rates of 0.25, 0.125, and 0.0625 resin-bed volumes per minute (rbv/min). The regenerant solution was divided into five portions, with four tanks out of five conserved for reuse. The fifth tank, which contained fresh regenerant, was pumped through the column at the end of the regenerating cycle. A résumé of the initial operating sequence used is shown in table 1.

Table 1. Initial operating sequence for pilot-plant strontium removal process

Influent	Flow rate *	Total gallons used	Temperature (° F.)
Tap water rinse-----	0.5	40	50
Milk-----	.125	200	50
Tap water rinse-----	.5	13.3	110
Wash (upflow)-----	.27	26.6	150
Tap water rinse-----	.5	26.6	70
Regenerant-----	.12	128	70
Tap water rinse-----	.5	40	70

* Resin-bed volumes per minute.

A series of seven runs, each lasting 4 hours at a pumping speed of 0.125 rbv, was made, and about 130 different bacteriological determinations were made on the milk and resin during each run. A summary of these results is shown in table 2. Between milk runs the wet resin was left in the columns. This was done to prevent resin packing, which results from drying the resin. Studies were made on the bacterial counts of the resin during storage at room temperature (approximately 75° F.) between milk runs. Growth of bacteria and yeast and molds occurred with the standard plate and psychrophile counts showed rapid increases (table 3). After storage periods beyond 7 to 10 days, mycelium was observed in the resin.

Table 2. Average percent increase in bacterial counts during the unmodified strontium removal process (pumping speed 0.125 rbv/min)

	Percent increase	
	Log average	Arithmetic average
Standard plate count.....	92	120
Psychrophile count.....	94	111

Table 3. Growth of microorganisms in resin during column storage (count/gram of resin)

	Storage interval (days)			
	0	3	7	9
Standard plate count.....	240	3, 900, 000	7, 500, 000	11, 000, 000
Psychrophile count.....	140	54, 000	210, 000	980, 000
Yeast and mold count.....	20	3, 000	4, 800	3, 200

Since it was feasible from a chemical standpoint to reuse part of the regenerating solution, we also were concerned with the microbiology of this solution during storage between milk runs. Growth of bacteria and molds occurred, although at a slower rate than in the resin (table 4).

In view of the rather rapid increases in the bacterial counts of the milk during processing and in the resin and regenerating solutions

Table 4. Growth of microorganisms in regenerant solution (count/ml)

	Storage interval (days)			
	0	3	15	25
Standard plate count.....	1, 600	2, 500	7, 000	30, 000
Psychrophile count.....	1, 700	2, 600	53, 000	350, 000
Yeast and mold count.....	33	500	800	2, 300

during periods of storage between runs, it was decided that the operating procedures used on the resin columns would have to be modified for sanitary reasons. Accordingly, laboratory studies were conducted to evaluate methods for sanitizing the column resin and regenerant solution. Columns with 1-inch diameter were used, with resin-bed depths of approximately 8 inches. The bactericidal and bacteriostatic effects of iodophors, chloramelamine, elemental iodine, acid, and hydrogen peroxide were studied. The laboratory studies showed that iodophors, chloramine T and chloramelamine dissipated rapidly when they were used to sanitize resin. Elemental iodine was excellent from a microbiological standpoint but severe off-flavors were produced in the milk when it was processed using resin that had been sanitized by iodine. Thiosulfate and sulfite rinses of the resin prior to running the milk did not solve the off-flavor problem. Table 5 summarizes the results of milk off-flavors from resin treated with elemental iodine.

Table 5. Taste panel results on milk samples from resin column sanitized with iodine

Minutes of milk flow prior to taking sample	Off flavor (number detecting/ number judges)
0.....	0/25
5.....	21/25
10.....	20/25
20.....	18/25
30.....	17/25

In addition to showing that iodine was useful microbiologically, the laboratory scale data showed that a combination of heat and pH would also produce satisfactory results. This technique involved a final resin rinse at 170° F. and then lowering the pH of the column resin to 2.0 by a rinse with a dilute solution of hydrochloric acid before storing the column. Regeneration of the column resin was done just before the milk run. These procedures were applied at the pilot-plant scale to see if they would be satisfactory microbiologically without affecting the taste of the milk when it was being processed. The operating sequence used in this phase of the study is shown in table 6. The combination of a 170° F. rinse followed by acidification of the column resin to pH 2.0 was found effective in preventing microbial growth in the resin so long as the heat treatment was repeated once every 2 weeks of resin storage in the columns (table 7). Every 2 weeks, heat treatment of the resin was used to prevent mold growth.

Table 6. Modified operating sequence for cationic radionuclide removal resin columns

Influent	Flow rate ^a	Total gallons used	Temperature (° F.)
Deionized water.....	0. 25	40	70
Regenerant.....	^b . 12	130	70
Deionized water.....	. 22	40	70
	. 0625		50
Milk (three alternates).....	. 125	200	
	. 250		
Tap water rinse.....	. 5	26. 6	110
Wash.....	. 27	53. 2	150
Tap water rinse.....	. 5	53. 2	170
Hot rinse.....	. 5	18	70
Acidified deionizer water.....	. 22	40	70

^a Resin-bed volumes per minute.

^b First 25 gallons were pumped through columns at a flow rate of 0.12.

Table 7. Microbial counts in column resin during storage at pH 2.0 (count/gram)

	Storage interval (days)			
	0	2	5	7
Standard plate count.....	< 10	< 10	< 10	< 10
Psychrophile count.....	< 10	12	< 10	—
Yeast and mold count.....	< 10	< 10	30	10

The laboratory studies also indicated that hydrogen peroxide in a concentration of 20 milligrams per liter at pH 2.0 was effective in preventing microbial growth in the regenerant solution. This technique was applied to the tanks of regenerant solution used for the pilot-plant process with excellent microbial control resulting (table 8). The residual hydrogen peroxide was reduced with sodium sulfite, and the pH of the regenerating solution was raised to 5.45 with ammonium hydroxide just prior to reuse for regenerating the column.

In addition to these modifications in operating procedure, a tubular heat exchanger was installed between the untreated milk storage tank and the resin column. This heat exchanger raised the milk temperature from that in the storage tank (35° to 40° F.) to 50° F. just before the milk entered the resin column. A 50° F. operating temperature for the column was advocated by the U.S. Department of Agriculture group

in the studies at Beltsville, because it was found that this temperature minimized the buildup of milk fat at the top of the column resin. The temperature of the treated milk was lowered to below 40° F. within a very few minutes after it left the resin column.

A series of 21 milk runs was made using (1) resin that was sanitized with heat and acid, (2) regenerating solution sanitized with hydrogen peroxide, and (3) a tubular heat exchanger to minimize the time the milk was above 40° F. during processing in the resin columns. Seven of the 21 runs were made at a pumping speed to process 200 gallons of milk in 2 hours, seven to process 200 gallons of milk in 4 hours, and seven to process the same volume of milk in 8 hours. The bacterial counts of the milk did not increase except after 6 hours of continuous milk processing (table 9).

Table 8. Microbial counts in regenerant solution during storage (count/ml)

	Storage interval (days)			
	0	2	5	7
Standard plate count.....	6	3	<1	<1
Psychrophile count.....	2	<1	<1	3
Yeast and mold count.....	1	<1	<1	2

Table 9. Change in standard plate count of milk during processing for cationic radionuclide removal

Total column running time (hours) for 200 gallons of milk	Percent change in standard plate count			
	Sampling time			
	2 hours (percent)	4 hours (percent)	6 hours (percent)	8 hours (percent)
2.....	-8			
4.....	-6	-12		
8.....	-5	-3	+4	+19

Since the sanitizing procedure for the resin and regenerating solution was based in part on acidification to pH 2.0, we studied the effect of this pH on type 304 stainless steel. Sections of 1½-inch milk pipe were put in jars of dilute hydrochloric acid at pH's of 0.5, 1.0, 1.5, and 2.0 and left in these jars at room temperature for 75 days. The section

of pipe left at pH 0.5 showed extensive discoloration and pitting, whereas only discoloration was noted in the section stored at pH 1.0. The sections of pipe stored at pH's of 1.5 and 2.0 did not show any visible effects. When the stainless steel lines and valves used with the resin columns were examined at the conclusion of the study, no discoloration nor pitting was found which was attributable to the acid used for column sanitation, with the exception of the lapped surfaces of the plugs of the three-way, hand-operated sanitary valves used at the bottom of the columns. These showed pitting after approximately 1 year of intermittent exposure to hydrochloric acid at pH 2.0. This pitting was not severe and did not affect the serviceability of the valves.

Throughout this study determinations were made of the possible effects that the changes which were made in the operation of the columns might have on the removal of strontium and on the taste of the milk. None of the changes made for sanitary reasons affected the removal of strontium. The sanitizing techniques developed for the resin and regeneration solution had no detectable effect on the taste of the milk being processed.

DISCUSSION

Question:

What was the average standard plate count of the milk used in this study?

Answer:

Our milk was obtained from farm-bulk-tank milk pickup trucks and the count varied from one load to the next. The average standard plate count was about 100,000 per milliliter.

Question:

Most of these studies were carried out where the column was stored for a matter of several days. Would you have the same recommendation for sanitizing the column on a day-to-day basis of operation? I think that, for day-to-day operation, hot water would hold the bacterial count down between runs.

Answer:

This procedure should be satisfactory for overnight storage of resin provided that there is not a buildup of thermophilic or thermoduric organisms. I have not demonstrated this, but in theory if you continue this technique for a period of time you are going to get a buildup of thermoduric and thermophilic organisms in your columns, and then your hot-water treatment will become progressively less successful.

Question:

When acid is used to store the column resin, is the resin regenerated before or after acidification?

Answer:

With acid storage the column must be regenerated after the acid is rinsed out, which is just before the milk run.

Question:

What was the composition of the regenerating solution?

Answer:

The regenerating solution consisted of sodium, potassium, magnesium, and calcium as chlorides in a concentration 10 times that in the milk being processed.

Question:

Did the use of pH 2.0 acid on the resin affect the amount of salt solution needed to regenerate the column resin?

Answer:

No changes were made in the amount of regenerating solution used after the acidification technique was developed. Since removal efficiency of the process was unaffected, it appears that no changes in regenerating solution volume were necessary.

Question:

Why were temperatures above 170° F. not used to sanitize the resin?

Answer:

The information available at the time of the study on the characteristics of the resin indicated that the resin might be damaged if temperatures higher than 170° F. were used. Since completion of this study, it has been found that higher temperatures could be used without adversely affecting the resin. One point should be emphasized. Even if temperatures of 210° F. are used to sanitize the resin, some bacteria will survive. These will grow rapidly and produce an unsatisfactory sanitary condition. Heat alone at temperatures that will not sterilize the resin doesn't appear applicable for sanitizing resin for extended storage periods.

DESIGN AND OPERATION OF FULL-SCALE EQUIPMENT

*H. E. Walter*¹

Dr. Todd:

Mr. Walter of the Department of Agriculture will discuss the operation of the full-scale equipment, using the reports we had this morning and findings from the laboratory to the pilot plant.

Mr. Walter:

This morning when Dr. Edmondson discussed the pilot plant, several questions were asked concerning potassium hydroxide. The particular reason we used potassium hydroxide for neutralization in the pilot plant was strictly a matter of flavor. We carried on pilot-plant studies using sodium hydroxide, potassium hydroxide, calcium hydroxide, and several combinations. Of all the combinations used, potassium hydroxide produced the best flavor in milk. A small percentage of sodium hydroxide can be added to the potassium hydroxide, but as far as the operation was concerned we felt it was better, even though the levels of potassium are increased in the finished milk samples, to omit the sodium hydroxide.

The design of the full-scale commercial plant for removing radio-strontium from milk is the prototype of the Beltsville pilot-plant system. As mentioned this morning, we originally used the 6-inch glass columns, operated by hand to control flow rates, batch acidification, and neutralization. Using this equipment, it soon became apparent that it was necessary to regulate and to sequence the operations properly to insure uniform strontium removal and to control the operation from a regulatory standpoint.

The present augmented Beltsville system was designed and submitted for bids and installation in 1963. Preliminary designs, plans, and specifications for the commercial system were established before the Beltsville system was completely installed. After installation, ex-

¹ Eastern Utilization Research and Development Division, Agricultural Research Service, U.S. Department of Agriculture.

perience with the Beltsville pilot-plant system suggested changes which were included in the final commercial design. Other changes resulted from observations in the Producers plant after we had the equipment installed for operations, from the recorded operational data obtained, and from the analytical work which was carried out.

Generally, increasing the capacity of equipment did not present any problems. As was mentioned this morning, the design was changed considerably, and certain pieces of equipment were eliminated. Originally, this was quite an automated plant, but to reduce the cost we had to cut out many regulator valves, reduce the number of regenerant tanks from four to two, and cut out all the in-place cleaning equipment. We did not make provisions for use of the vacuum pan to remove the amount of water added to citric acid and in the potassium hydroxide.

Now, in going through this matter briefly, reference is made to the Preliminary Report of the Full-Scale System, dated October 1963, previously distributed. Let us discuss this system first. Acidification (fig. 1) is probably one of the most important parts of this process. As shown this morning, the degree of acidification determines, to a great extent, the amount of strontium removal. At pH 5.2 to 5.3 under controlled conditions, 95 to 98 percent of the strontium is available for removal.

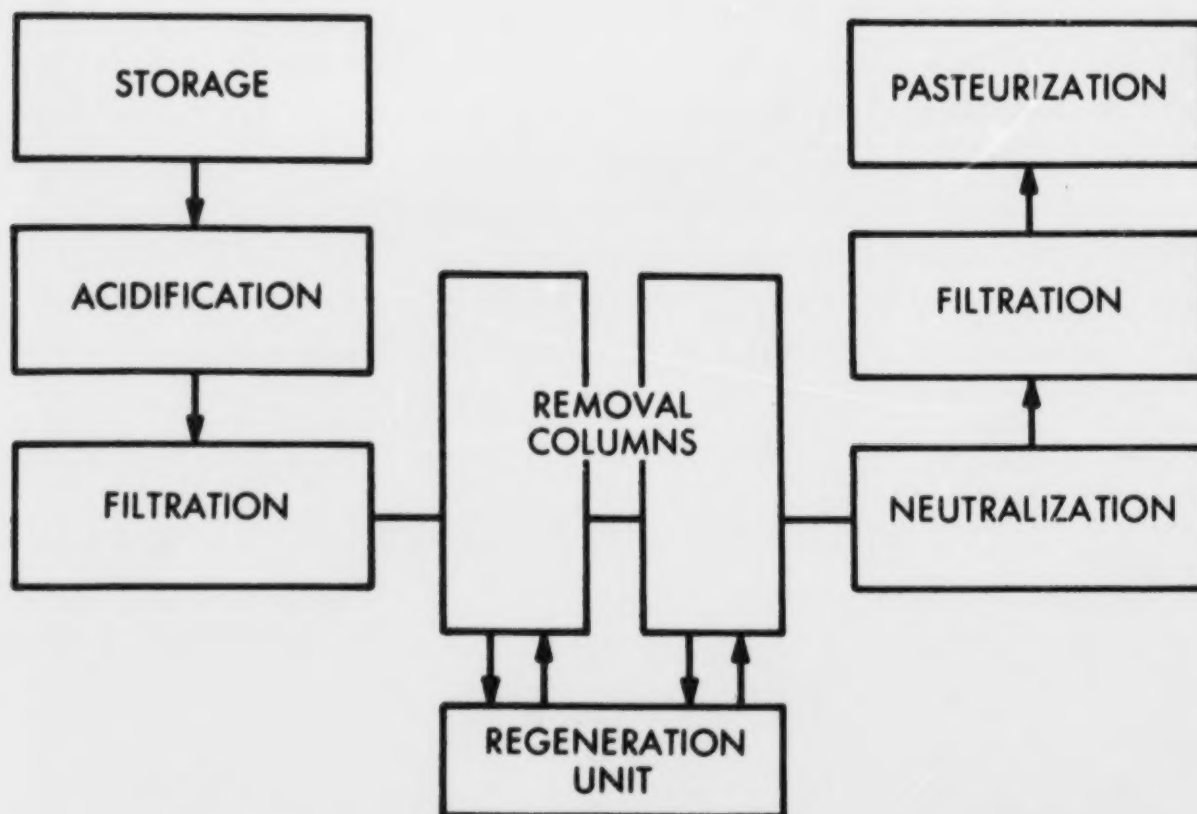


Figure 1. Removal system for strontium from milk

In our original pilot-plant work we injected the acid through a small orifice directly into the restricted milk line. This produced satisfactory results on a very small scale, but it soon became apparent that this could not be done on a large commercial scale. At that particular time, sanitary, commercial acid milk-mixing units were not available. Mixing units were available, but they were not constructed to provide complete breakdown for cleaning. The Producers Creamery personnel then developed an in-line centrifugal pump with the acid injection stream impinging under pressure directly on the rotor. We have had little trouble, if any, with localized precipitation.

Last Sunday we processed 104,000 pounds of milk through one filter. Pressure buildup, if I remember correctly, increased from 28 to 34 pounds, a 6-pound increase throughout the entire operation. You can see there was little, if any, actual precipitation from the lack of any great pressure increase. The pH for the most part ranged between 5.25 and 5.35.

On the acidification chart (fig. 2), you will notice that after acidification there is a pH sensor. Around that pH sensor is a bypass. This bypass is quite important because you have to acidify this milk and circulate the milk back to the original milk storage tank until the proper pH is attained. If you tried to put the milk through the column at a pH of 5.7, 5.8, or 5.9, it would be apparent in the results that you have removed not more than 75 percent of the strontium. The milk is pumped through the pH sensor to actually record the pH of the milk on the pH recorder. With a laboratory pH meter which we had run previously, the pH of the milk was 6.7. At that point, the pH is established on the recorder at 6.7, so that we know the pH of both meters correspond. The acid pump is then turned on and the acid is injected until the pH of the milk drops below 5.4, and this, in turn, is checked on the laboratory pH meter so that we have a double check as to the exact pH. This particular pH sensor in the Producers plant is the result of development work on sanitary pH units by Producers personnel, U.S. Department of Agriculture, and Beckman Instrument Corporation. The units that we have in Beltsville are stainless-steel sensing units, but they were not designed to comply with strict dairy standards. The particular pH sensor that you will see tomorrow is constructed almost entirely of standard fittings, 3-A.

After the milk has gone through the pH sensor it passes through the filters. The reason for this is to recover all of the precipitation that occurs. If we do not have the filters and get this localized precipitation in the column, it is exceptionally difficult to get the precipitate out of

the resin. For those of you who have had experience, it is a lot of fun to make cottage cheese, but not in an ion exchange column.

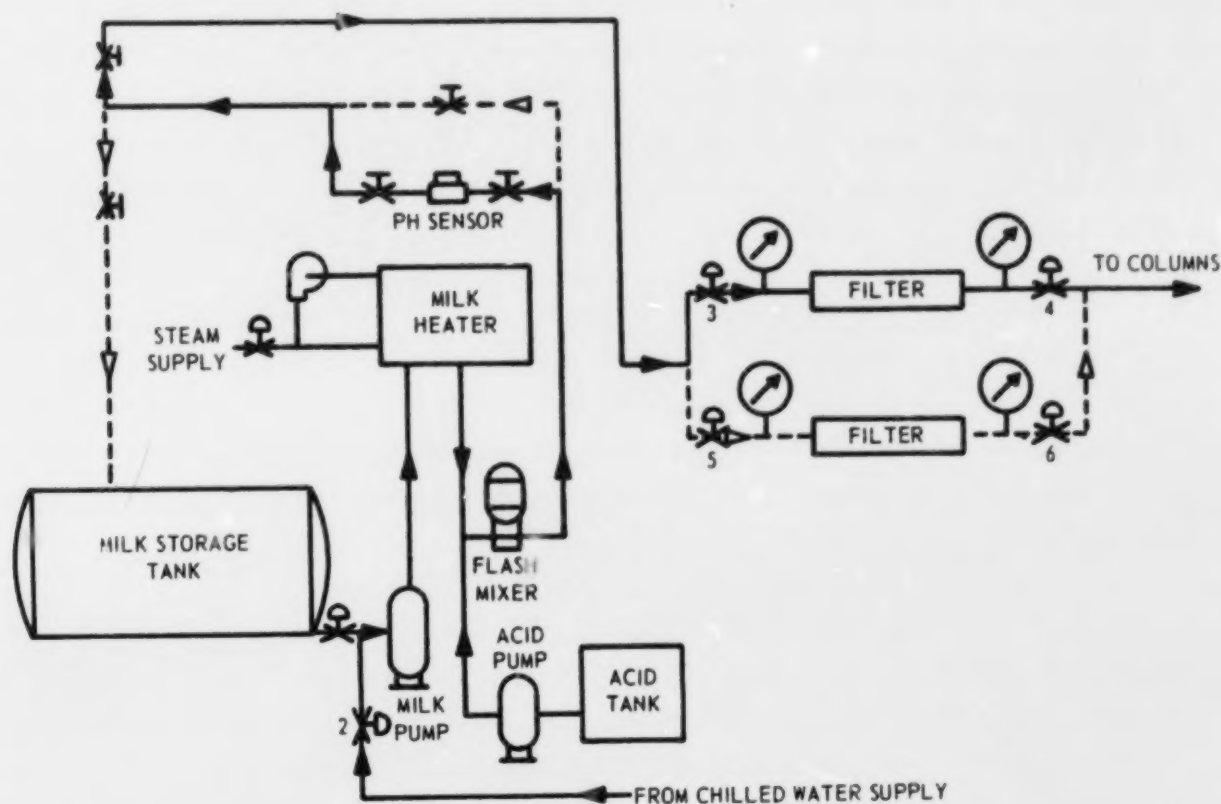


Figure 2. Acidification

The reason for the two filters is to maintain continuous flow. If you are injecting this milk at the rate of 12,500 pounds an hour and a precipitation develops, you have a pressure differential in the system so that it will automatically switch from one filter to the other; otherwise, you would have too much pressure buildup.

These filters also remove some butterfat. The colder the milk, the more butterfat will be removed from the milk in these filters.

The columns that we used in our pilot plant at Beltsville, as Dr. Edmondson said this morning, were 11½ inches in diameter and about 80 inches tall. These columns had a ratio of about 7 to 1. The column ratio in Producers plant is about 2¼ to 1. This was necessary because of the ceiling heights of the buildings. Their columns are 3½ feet in diameter and about 8 feet tall.

The milk flow and removal efficiency have been satisfactory in the columns here at Producers. However, we have had problems in back-washing the resin or actually cleaning the resin. The columns we had

at Beltsville were designed so that the screen, or the perforated plate at the bottom of the column, was exactly the same diameter as the column itself. Therefore, when the water was pumped into the bottom of the column, there was a complete wash of the entire surface. In the large columns we had a terrific amount of difficulty in trying to figure out how to put the screen in so that it would hold the resin properly and yet would be removable. So, in these columns we had a perforated plate with a 2-inch rim next to the inside periphery of the column. As a result, pumping backwash into the column to wash the resin developed a pocket between the sidewall and the resin screen. During our washing operations cold and hot spots could be detected on the outside of the column surface. We knew it was not circulating as it should be.

During the backwash operations, even though we had high-capacity pumps up to 80 gallons a minute, there were still blind or cold spots on the column surface. To control this, Producers people installed an air-sparging unit to see if we could disturb the resin with air plus the backwash to maintain complete washing. This helped a great deal. However, on the run of October 15, the combination of 80 gallons of water per minute with a few pounds of air pressure ruptured the screen at the top of the column. We had much better flow after that. You will see the ruptured screen tomorrow at the Producers plant. I am not saying that if we did not have this 2-inch rim (in the commercial columns) we would have had perfect conditions—I don't know. I am just comparing the system at Beltsville with this system. The plate that we used at the bottom of the column was a photo-etched stainless steel plate. In Beltsville, we originally used stainless steel screening. Of course, that is difficult to clean, so we wanted to use this particular plate. It is very thin; the small holes are 0.012 inch in diameter. Each column is also equipped with an air bleed valve. For example, there were occasions when we had to drain the water from the column, and there must be an air intake so that the column can be bled.

During operations in the commercial system, as I mentioned, the column is 8 feet high, our bed volume was 5 feet (350 gallons of resin in a column) so we had approximately 3 feet of expansion. To use this in this head space, our system was flooded. Flooding this system with water or with milk is necessary for several reasons. If there were no flooded head system on our particular design, for one thing, the milk flowing into the column would channel the resin which would reduce the distance of flow through the column; besides, it would provide, with this particular head space, more time for the milk to become

equilibrated after acidification. This head space would accommodate well over 300 gallons. You see, this is almost the same as a bed volume. Thirdly, it provides better milk control.

We tried at Beltsville on numerous occasions to operate with a head space half full, and of course, developed pressure problems in controlling flow.

After the milk has passed through the resin column itself, it is neutralized with potassium hydroxide (fig. 3). On the first few runs at Producers we did not have the centrifugal mixing pump; we used an in-line injection directly from the neutralizer pump. We had a very erratic control. It was exceptionally difficult to control the pH accurately on neutralization. As a result, we soon put in the centrifugal mixing pump on neutralization, and we had much better overall control.

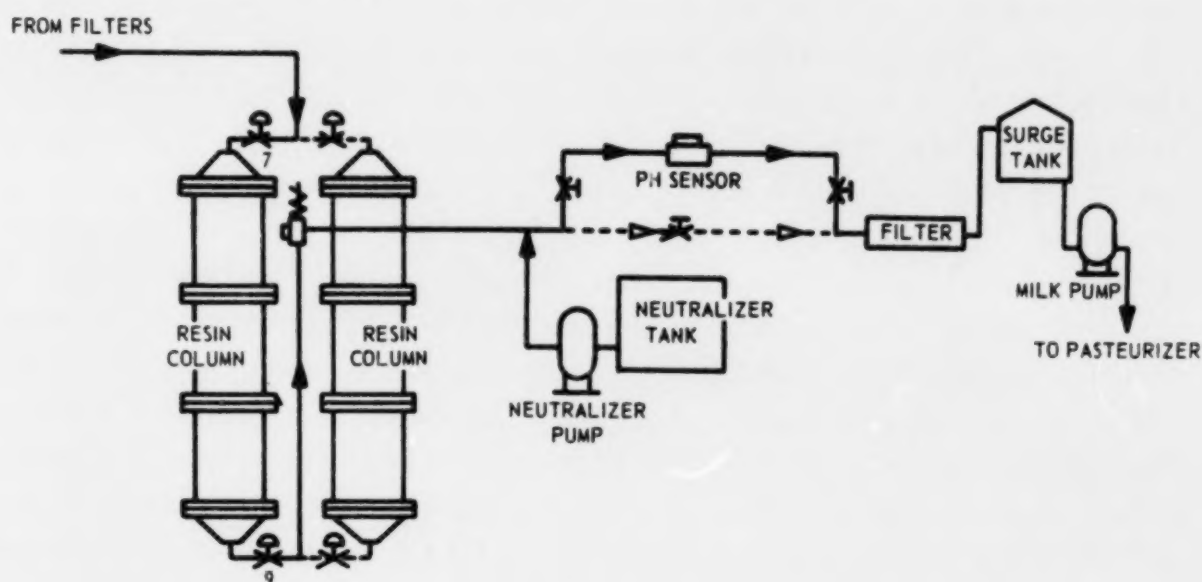


Figure 3. Ion exchange and neutralization

In addition to the pH sensor, there must also be on that system a pressure differential gauge to maintain pressure in the column. If there isn't, there is a buildup of pressures of 25 to 35 pounds going into the columns, and flow rates cannot be controlled. Sample cocks were installed in many places and used primarily for bacteriological and pH control samples. Samples for the laboratory pH were taken every 2 to 4 minutes, originally, until the desired pH was established, then every 15 to 30 minutes, and checked on the laboratory pH machines.

After the milk has gone through the pH sensor at the end of the neutralizing system, a filter is necessary to retain the glass, should the glass electrode break, or any resin that might come through the column screen.

Originally, we used a small 50-gallon surge tank. We soon found that it would not operate correctly on a large system, particularly when the KOH injection wouldn't work properly and the pH of the milk dropped to 6 or below. Milk at pH 5.75 heated to pasteurizing temperatures resulted in a large amount of "burn-on" in the pasteurizer. It was necessary to use a large surge tank to maintain and stabilize proper pH control.

With the large surge tank containing 6,000 to 15,000 pounds of milk, we actually used one of the large 5,000-gallon bulk tanks. This provided an uninterrupted flow of milk to the pasteurizer at pH 6.75 to 6.85. This morning we mentioned pH 6.6. In all cases, the pH of the raw milk hasn't been above 6.7 so we neutralized only to pH 6.6. This milk had a pungent taste and in the runs since then we have been neutralizing to pH 6.75 to 6.8. Much better quality and better flavored milk resulted when the milk was neutralized to the original pH or slightly higher. (This was the judgment of the taste panel in Washington as well as the Producers panel.)

One of the greatest problems that we had, other than the ion exchange work itself, was the burn-on in the pasteurizer. This burn-on has not been in the heating section or pasteurizing section; it has been on the regenerant side of the pasteurizer section. The result of this burn-on was that we had to reduce the overall capacity of the system to maintain temperature within the pasteurizer itself.

Since we used the large surge tank we have had much less burn-on. The run tomorrow is being made to find out if we can actually process this ion exchange treated milk through a pasteurizer for a period of 16 hours. It has been processed many times for 8 hours, but it is absolutely essential that we run a 16-hour pasteurization to see if we could actually pasteurize that milk without a large burn-on on the pasteurizer itself.

As the water temperature would be increased to maintain temperatures in the pasteurizer, the burn-on, of course, also increased. We don't know exactly what this burn-on is. It appears to be a proteinaceous material. The run of February 21 had the smallest amount of burn-on that we have had at any one particular time. We do not think it will be a problem today. We feel now that we can operate for 16 consecutive hours.

In the actual system itself, it is necessary to use agitation to mix the acid and the KOH, and then to maintain agitation not continuously, but several times during the day's operation.

Agitation of the regenerant solution during mixing is very important. It must be maintained so that all salts are completely dissolved. With the water at Producers Creamery Company we developed a flocculent condition in the regenerant. At times this would settle out in the resin column itself. With the addition of hydrochloric acid to pH 5.35, the regenerant would be just as clear as water, and we had no further problem.

In the past we have been using single-pass regenerant. As I mentioned previously, one of the results of cutting down on the overall cost was not using the four regenerant tanks. Consequently, we have used 14 bed volumes of regenerant solution in two tanks. From the data that Dr. Edmondson gave this morning, by increasing the regenerant temperature of the last three 8-hour runs, only nine bed volumes of 140° F. regenerant will be required. From that data we feel that we can strip the strontium from the resin and at the same time regenerate properly. No doubt other speakers this afternoon will suggest changes to improve this system, both from engineering and sanitation standpoints.

The citric acid that we used was made up with warm water. Dr. Marshall set up titration values to titrate the citric acid and the KOH. We had problems originally with the large 500-gallon KOH tank. Don't be surprised tomorrow when you see the boat paddle we use to mix the potassium hydroxide. Mr. Sparling had quite a time justifying the purchase of a paddle for a strontium 90 project, but in lieu of a mechanical mixer it worked better than anything we had.

The citric acid solution made up with warm water is 19 percent, using 1.6 pounds of citric acid for each gallon of solution required. There is very little variation in the amount of acid injected; the control does not vary more than $\pm .01$ pH. It is very accurately controlled.

The more you increase the strength of acid or KOH, the more difficult it is to control. By changing the concentration, we were able to get a fairly even KOH control.

Originally, we were using resin regeneration the same day in which we processed milk. This was primarily because we sent our strontium 90 samples to Boston for analysis, but we soon found out that it was far better to hold the resin itself in a clean or washed condition rather than in the strip condition. In our particular work here, of course, the columns were washed every few days to try to maintain sanitary conditions.

We have been trying to maintain, as much as possible, a comparatively slow regenerant flow rate with the resin concentration. The figures that we used here for 1 resin-bed volume of regenerant were 117 pounds of calcium chloride, 60 pounds of potassium chloride, 28 pounds of sodium chloride, and 54 pounds of magnesium chloride.

Some of the associated problems relating to central automation and quality control will be discussed by the other speakers. All of the equipment problems have not been solved up the scale from the pilot plant to the large-scale operation. Additional large-scale operations will be necessary. However, Producers research has shown that this operational, large-scale system is both feasible and practical.

Certainly the system would be designed differently now that we have gained the experience of 2 years of research. I think that the success of this commercial research project has been due primarily to the tremendous efforts and cooperation of the personnel in the Producers Creamery Company. Thank you very much.

DISCUSSION

Mr. Walter:

Are there any questions or comments?

Dr. Murthy:

Were CP chemicals or commercial grade used?

Mr. Walter:

The only one that was commercial grade was the calcium chloride, and that was equivalent to, but not CP grade.

Dr. Hoover:

I don't believe that all the people might have understood the explanation of why you were considering potassium for neutralization. It must be realized that when you neutralize with potassium, the milk that comes out of this system has considerably higher radioactivity than it had when it went in. If you have 1 part in a billion of strontium that is radioactive, you don't have much; whereas, 1 in 600 of potassium is radioactive, and you do have a much higher level of potassium present. When you increase that about 68 percent, you are increasing the radioactivity as measured in milk quite significantly. It can be argued you should not do this. However, the body does not retain the potassium as it does strontium.

PROBLEMS ASSOCIATED WITH EQUIPMENT DESIGN

*J. H. McCutchen*¹

Mr. McCutchen:

I am going to tell you a bit about the sanitary design and the problems we had in relation to this equipment. After Dr. Hoover, Mr. Terrill, and other officials decided to build this equipment and gave us the money to buy the hardware, it came down to the matter of expediting and building the equipment, getting it into the plant, and getting it to operate as designed. When this occurred, they called in the second team, and I was on this second team to get the hardware set up. We had a number of conferences with personnel of the Paul Mueller Company and the Producers Creamery Company, and looked over several sets of plans and specifications to get the bugs worked out of the equipment.

At the first meeting, I told them that I had a rather simple request to make as far as sanitary design of the equipment was concerned: All I wanted was equipment that would meet 3-A Sanitary Standards.² Equipment which had no 3-A Standards should meet the U.S. Public Health Service Grade A Milk Ordinance and Code. This, of course, caused ulcers among the equipment-manufacturing people because they had to produce this sort of equipment and some items had never been designed or built before on this scale.

As a result, I made several trips to the Paul Mueller plant while the equipment was being fabricated. We cut through some of the welds on the equipment and cross-sectioned some of the things to be sure that we were getting the proper welding and proper finishing on the equipment that was being assembled. We designed some features for this equipment that we would not put on regular milk production equipment.

¹ Division of Environmental Engineering and Food Protection, Public Health Service, U.S. Department of Health, Education, and Welfare, Present address: Environmental Sanitation Program, National Center for Urban and Industrial Health, PHS, U.S. DHEW, 222 East Central Parkway, Cincinnati, Ohio 45202.

² 3-A Standards Jointly Promulgated by the Sanitary Standards Subcommittee of the Dairy Industry Committee, the Committee on Sanitary Procedure of the International Association of Milk and Food Sanitarians, and the Milk and Food Program, Division of Environmental Engineering and Food Protection, U.S. Public Health Service.

We put in sight glasses and sampling cocks because we didn't know in this type of research operation just what kind of tests would be necessary, and just how much visual operation we would want to have during the process. When you examine this equipment tomorrow, remember that this is a research type of operation and these features would not necessarily be provided on regular commercial milk equipment. We have gone as far as we can to make this equipment meet as many of the sanitary designs, standards, and requirements as possible. In some cases, the equipment was not available in sanitary design, and in other cases it was built from scratch, you might say, to meet this need. But we feel rather proud of the job that has been done, and feel that the equipment installed at the Producers Creamery Company plant complies, in most respects, with sanitary design standards.

DISCUSSION

Mr. Terrill:

I would like to ask what the lead time would be—in your judgment and perhaps the judgment of others—in case somebody would want to duplicate this kind of equipment. What sort of lead time would have to be planned?

Answer:

I think that some of the equipment manufacturers could answer this question. Is anyone here from the Paul Mueller Company?

Could you give us some idea on how long it would take to build this type of equipment from the beginning?

Answer:

Four to 6 weeks for tanks, pipe, and columns.

Question:

Would this include the automated controls too? I ask this because I know how long it took to get the first set.

Answer:

No, the automated controls require a lot longer period of time—from 3 to 6 months.

AUTOMATION OF FULL-SCALE SYSTEM

*Eugene M. Sparling*¹

Mr. Sparling:

Perhaps the most logical place to start this discussion is to review the present system. The schematic is from the raw milk storage tank through the resin columns and related equipment to the storage tank, which is used as a surge tank before the high-temperature short-time pasteurizer. The HTST pasteurizer system is of conventional design, and therefore familiar to all of you. The system is controlled by two Taylor Instrument Company Flex-O-Timer controllers and Flex-O-Steppers working in harmony, one Flex-O-Timer and one Flex-O-Stepper combination for each resin column. In the future they will be referred to as timer-steppers as a matter of convenience. The timer-steppers are interlocked so that one timer-stepper combination cannot be activated while the other column is operating. The timers activate air valves, stop and start pumps, and turn off and on heat and pH control systems at a predetermined time. Actual timing of the milk through the resin column is accomplished by means of the Taylor Auxiliary Timer with a 15 minute to 12 hour time range (fig. 1).

The pH control is presently accomplished by two separate but basically alike systems; one system for the addition of acid, and one system for the addition of base. Each system relies on the Beckman Model J Analyzer, range 0 to 10 pH for sensing pH, coupled with a Taylor Transcope Recording Control Station with Integral Auto-Manual Timer Unit, set point transmitter with plug-in control and alarm to record and control the acid and base additions to the milk. This system must be checked regularly by laboratory pH meters to assure complete accuracy. The acid is metered into the system by means of a Milton Roy Pump with a maximum capacity of 39 gallons per hour at a 145-pound discharge pressure. The base additions are by a Milton Roy Pump with a maximum capacity of 95 gallons per hour at a 120-pound discharge pressure.

¹ Producers Creamery Company, Springfield, Mo.

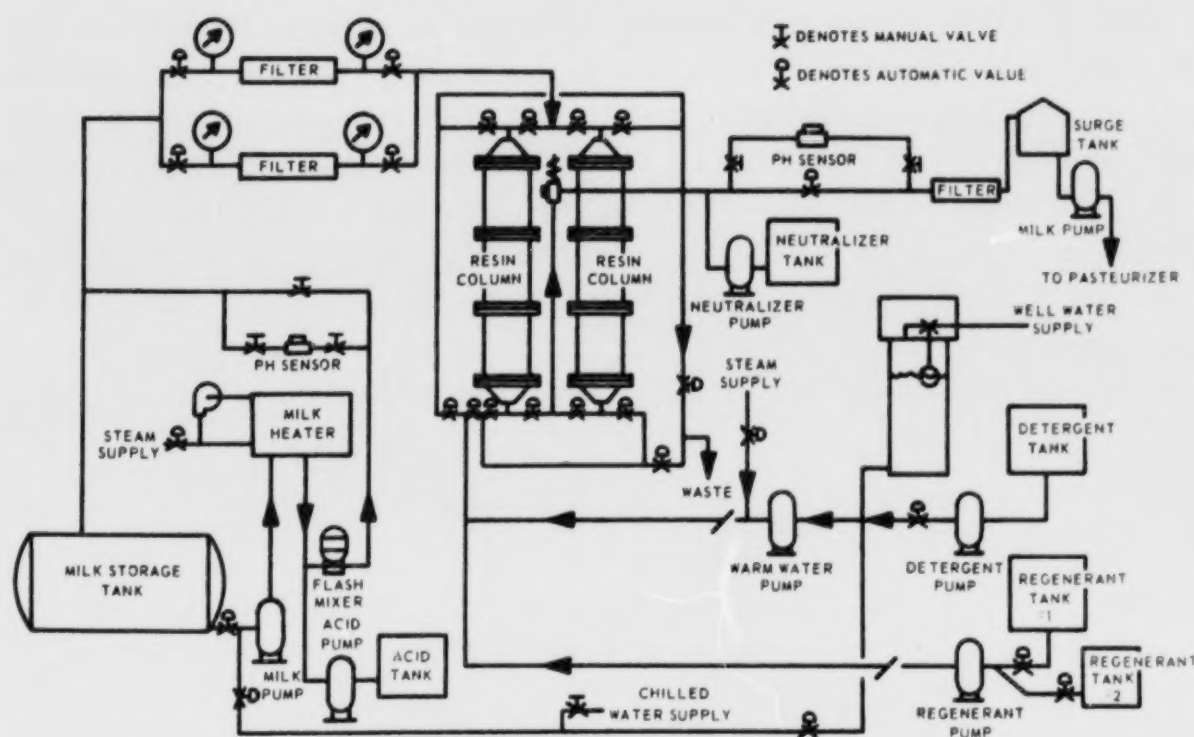


Figure 1. Removal system for Sr from milk. Schematic flow diagram

The following is a description of the program on the timer-steppers. The initial condition, when air and power are on, finds all valves closed except CV 25 and 19 or CV 24 and 18. Essentially this means that one set of filters is open and ready for forward flow. At this point either resin column can be activated at the discretion of the operator. We will assume that column No. 1 is chosen. The auxiliary timer, which controls the length of time the milk passes through the resin column, is set to the predetermined number of hours chosen for the removal portion of the run. The start button is then pushed to activate the system.

Step 0: This is the time when the timer-stepper scans the controls to determine the status of No. 2 control system. If the No. 2 control had been activated and was in any of the sequences prior to step No. 8, the No. 1 control system would go into a ready-wait condition. When the No. 2 stepper-timer cycles to step 8, the No. 1 timer-stepper proceeds into step 1.

Step 1: The interlock goes on so that column No. 2 cannot be activated into forward flow. Refrigerated water starts through the system from a point between the raw milk storage tank and the raw milk timing pump to temper the resin column and equipment to a temperature of 55° F. or cooler. For whole milk, the temperature preferably should not be below 50° F. to avoid excessive hardening of the butterfat.

Step 2: This is the pH sensing adjustment period. Milk from the storage tank is started into the line through the preheater, through the pH sensing system, and returned to the milk storage tank. The acid addition system is not activated. This period is devoted to standardizing the acid pH meter in a moving medium against a laboratory pH meter. The preheater is activated and controlled to 55° F. The plate preheater is of conventional design. During this period the resin column is precooled by transferring the water supply from the raw milk timing pump, P.-1, to the resin column wash pump, P.-4.

Step 3: This is acid control. The acid pump and the line mixer, LM-1, are activated, and the pH of the milk is standardized to between pH 5.3 and pH 5.4.

Step 4: The milk is manually diverted from the recirculating condition to forward flow. Pump 4 is stopped and appropriate control valves are changed to allow the milk to flow through the resin column. At this time, column No. 2 can continue into any step after step 8 if it is so activated.

Step 5: The auxiliary timer takes control of the system and holds the timer-stepper in step 5 for the elapsed time preset on the auxiliary timer. After the start button has been activated, the only control over the auxiliary time is to shorten the time span. The base control system is activated; and during the first part of this period, the pH sensing device is standardized, and the sensitivity of the control is checked.

Step 6: This step is activated at the conclusion of the time set on the auxiliary timer. Refrigerated water is introduced into the system at control valve 22, and the milk is rinsed from the system. The acid-metered pump is turned off.

Step 7: The preheater is off. Rinsing is continued through control valve 22 for a sufficient time to drive the milk from the resin column. It is assumed that the 15 minutes allowed by steps 6 and 7 are sufficient time to accomplish this function.

Step 8: Base metering pump is turned off; the interlock is off. Column No. 2 can proceed into step 1 if it has been activated. Column No. 1 returns to initial conditions for 30 minutes to allow column No. 2 to pass through the first four steps.

Step 9: Start to rinse the column through pump 4, in forward flow; effluent is directed to waste. The wash-water temperature is controlled by a Taylor Full-Scope Recording Temperature Controller, TRC-2, which controls an air regulated steam injector. The desired temperature is manually set.

Step 10: The wash-water flow is reversed through the resin column; that is, it enters the bottom of the resin column and exits through the top. At this time the pump is delivering approximately 75 gallons of water per minute to expand the resin to allow more complete washing. The detergent auxiliary timer is activated, and in turn activates, times, and deactivates the detergent pump and control valve. The time on this auxiliary timer should not exceed the time allowed for this step, since the timer has no power to extend the step past the time interval set on the timer-stepper.

Step 11: The rinse-water flow is changed to enter the resin column at the top and to exit from the bottom. The detergent timer is off.

Step 12: The regenerant pump starts, and appropriate valves open; regeneration is in forward flow.

Step 13: This is the same as step 12 only regenerant tanks are changed.

Step 14: Rinsing is started in forward flow at the same rate of flow as regeneration for the first 10 minutes; after that the rate may be increased, if desired. The temperature is increased so that the water exits from the resin column at 180° F. or hotter to sanitize the resin.

The last step is a return to initial conditions.

It has been mentioned earlier that regeneration is now accomplished immediately before the run. It is manipulated manually as a matter of convenience. In the case of daily use, regeneration would be controlled by the timer-stepper. At present, the high-temperature, short-time pasteurizer is programmed by the timer-stepper to start step 1 and to turn off at step 9. This was not mentioned during the discussion of the steps because it was found necessary to operate the unit more independently than is possible when it is controlled by the timer-stepper. For this reason the high-temperature, short-time pasteurizer is started and stopped manually.

You will also note that wash pump P.-4 is operated at various flow rates. These flow rates are changed manually.

In the present process, one of the critical points is milk flow through the resin bed. This flow rate varies through the run owing to back pressure developed in filters 1 and 2 and in the resin bed. For this reason, control would be improved by placing a flow meter in the line before the resin column to allow accurate calculations of flow rates during the run.

The pH control and acid and base metering were initially a problem. Beckman sensing devices were erratic at first and not completely

dependable. It is now felt they are relatively dependable, but do need, and probably always will need, laboratory backup. The Milton Roy pump is a single-piston pump that gives a pulsating feed. The Milton Roy pump is hypersensitive to moisture and oil in the air that supplies the control mechanisms. We have been informed that engineering changes have been made to remedy this problem. It is felt that the pH control may be better accomplished by using a constant-speed, proportionate feed system. The line pH meters would then be used to monitor and not to control the pH. The pH control would be accomplished by hand regulation of the acid and base feed by means of air-operated flow control valves or by constant feed variable speed pumps.

The control valves used on the project are Grinnell-Saunders, straightaway diaphragm valves with black butyl diaphragm, and Taylor Air-Activated Actuators. Some problems are experienced by operating the valves in pairs. When one valve is closed and the other is opened simultaneously, the rate of opening is not uniform because the valves are opened by spring pressure and closed by air pressure, which may result in both valves being closed at once. Air to open and close valves or stronger opening springs are indicated. Diaphragms did not hold up under the service to which they were put.

In evaluating the present system we find the air-activated control valve should be placed in the line at the point where the milk flow is diverted back to the raw milk storage. These valves should be connected to the pH alarm system on the acid side and automated to divert the milk flow back to the storage tank when the pH becomes either too high or too low. This would protect the system from being laden with precipitated proteins from overacidification or from poor strontium removal in the resin column because of underacidification. Air valves were in the original plans, but in the quest for economy they were replaced with manual valves.

In the present system the control valves CV 1 and 5, 4 and 8, 2 and 6, 3 and 7 are programmed in pairs. This was done so there would always be valves opened to and from the resin column so that excessive pressure would not be exerted by pumps on the columns. However, if these valves had been programmed separately, the present system could have been automated to change resin columns during operation without disrupting the flow of milk. This would be accomplished by closing the milk input valve on CV 2 or 3 on the retiring resin column, and opening the water input valves 1 or 4 and CV 9 and starting pump 4. At this same time the reverse would be occurring on the column just entering service. The retiring column would continue to feed milk from the exit

of the column into the system until the water starts diluting the effluent. During this period the water draining from the fresh column would be diverted to waste through the control valves at the bottom and CV 13. At the time the water appears in the effluent of the retiring column, milk would appear in the effluent of the entering column. At this time the effluent of the retiring column would go to waste, and the effluent from the entering column then would go into the system. The retiring column could then be rinsed, washed, and regenerated, and be ready for return to service if needed. This could go into a continuous process where the two resin columns could then be rotated every 8 hours.

The Flex-O-Timer could be discarded. You may remember that in early steps of the present program the timer was sequenced to allow manual adjustments to the pH meters in preheated condition. The time required to standardize the line pH meter and then standardize a moving product to a given pH is not totally predictable. We find instances when the timer must be stopped to allow more time for adjustment. The same holds true at the end of the cycle. The time required to rinse and wash a large volume of resin clean is not totally predictable. This is true of other operations in the process. Since there must be an operator present constantly monitoring the process, total automation at this time would be a handicap because the operator must be ever mindful of the time for each step in relation to the manual adjustments that are part of the function. These steps might be activated by a series of buttons or by a manual dial up to step 5, when the auxiliary timer takes over and times the duration of the milk run through the resin columns. After the auxiliary timer has run its course, the button or dial system then would be used to continue sequencing.

The present pressure-sensing arrangement on filters F-1 and F-2 is highly recommended. This control is set to a predetermined pressure. When the line pressure ahead of the filter reaches a preset pressure, the controller changes filters and sounds an alarm so that the operator may change the filter discs and place the filter back on the line in a standby condition. The controller is reset so that the next time the pressure reaches the preset pressure the flow will be diverted back to the first filter and the process of changing filter discs is repeated.

PROBLEMS ASSOCIATED WITH QUALITY CONTROL AND REMOVAL EFFICIENCY AND PROBLEMS ASSOCIATED WITH PROCESSING

*B. R. Heinemann*¹

Mr. Heinemann:

As Dr. Hoover indicated this morning, the first run at this plant was made on May 22, 1964. Additional runs were made on June 24, August 20, September 16, and October 14. Although three additional runs are now in progress, the results are not available, so this report will be confined to the six full-scale runs which were made during 1964. At the completion of the three current runs, the plant will have been operated on milk produced during all seasons of the year.

For each run the milk was shipped in tankers to the plant, each tanker holding about 35,000 pounds of milk. Samples were taken from each tanker, from pipelines as the milk was pumped to the columns, from the pipelines after leaving the columns, and from the processed pasteurized milk in the tankers as they left the plant. The milk was then taken to our Monett plant where it was dried and sold for animal feed. A minimum of about 40 samples were taken from milk during each run, and approximately 200 different chemical and bacteriological tests were made. All determinations were made in duplicate or in quadruplicate. On the basis of these determinations we will discuss and evaluate the results of the radiological data, chemical determinations, microbiological results, and organoleptic changes.

We will discuss first the strontium 90 removal. Table 1 presents the strontium 90 removal efficiency for all six runs. In order to have the results of the operations of the columns as quickly as possible, a rapid test was used permitting results to be obtained in 12 to 14 days.

These results are referred to as the rapid test. The conventional procedure required approximately 8 weeks to perform, and these results are presented in the table as long-test results.

¹ Producers Creamery Company, Springfield, Mo.

Table 1. Removal efficiency of ^{90}Sr as determined by both rapid and long tests for ^{90}Sr

Sample	Skim milk run 143		Whole milk run 176		Skim milk run 233		Whole milk run 246		Skim milk run 260		Whole milk run 288	
	Rapid	Long	Rapid	Long	Rapid	Long	Rapid	Long	Rapid	Long	Rapid	Long
Raw-----	39.0	42.2	38.4	36.0	31.5	32.9	37.9	43.7	32.4	31.0	25.5	25.6
5 BV-----	2.2	2.3	1.3	1.1	2.4	1.2	3.1	2.1	2.8	1.5	5.1	4.1
10 BV-----	2.1	2.2	1.8	1.1	2.6	1.3	2.4	1.9	6.7	5.7	3.0	2.8
15 BV-----	2.3	2.0	3.2	1.1	2.9	1.2	3.1	1.5	2.7	1.6	2.6	2.1
20 BV-----	2.7	3.2	1.0	1.2	2.7	1.7	3.2	2.4	1.7	0.9	2.7	2.2
25 BV-----	3.9	2.8	1.2	1.3	2.4	1.5	2.7	1.7	2.3	1.4	3.1	2.8
30 BV-----	5.5	5.0	1.4	1.0	4.0	2.5	3.4	2.2	3.4	2.4	2.6	2.8
Processed-----	•16.6	•12.4	2.0	1.1	3.1	1.8	3.5	2.4	3.1	2.1	3.2	2.6
Average of line samples-----	3.11	2.90	1.65	1.13	2.83	1.57	2.98	1.97	3.27	2.25	3.19	2.80
Percent ^{90}Sr removed-----	92.0	93.1	94.8	96.9	90.2	94.5	91.8	94.5	90.4	93.2	87.5	89.9

Percent

Average removal for six runs, rapid test-----

91.12

Average removal for six runs, long test-----

93.68

pCi/liter

Average raw samples, rapid test-----

34.12

Average raw samples, long test-----

35.23

Average processed samples, rapid test-----

3.00

Average processed samples, long test-----

2.15

Average line samples, rapid test-----

2.84

Average line samples, long test-----

2.10

• Results excluded from calculations.

As shown on the table, the rapid-test results compare very favorably with the results obtained by the long-test method. The average for the raw milk samples determined by the short-test was 34.12 picocuries per liter, while the long-test result was 35.23 picocuries per liter. Thus, for control purposes, the rapid-assay method would prove to be entirely satisfactory.

You will also note from the table that the average of the line samples checked out very well with the average of the composite samples taken from the milk after processing. This table also shows that the removal efficiencies from the various runs ranged from 89.9 percent removal on run 288, the last run made, to a high of 96.9 percent on run 176. The average removal efficiency by the long-test method is 93.68 percent.

As shown in figure 1, every fifth volume of milk was sampled and the strontium 90 content determined on that sample and then averaged for the entire six runs. It will be noted that the strontium 90 content is very uniform throughout the 30-bed volumes sampled.

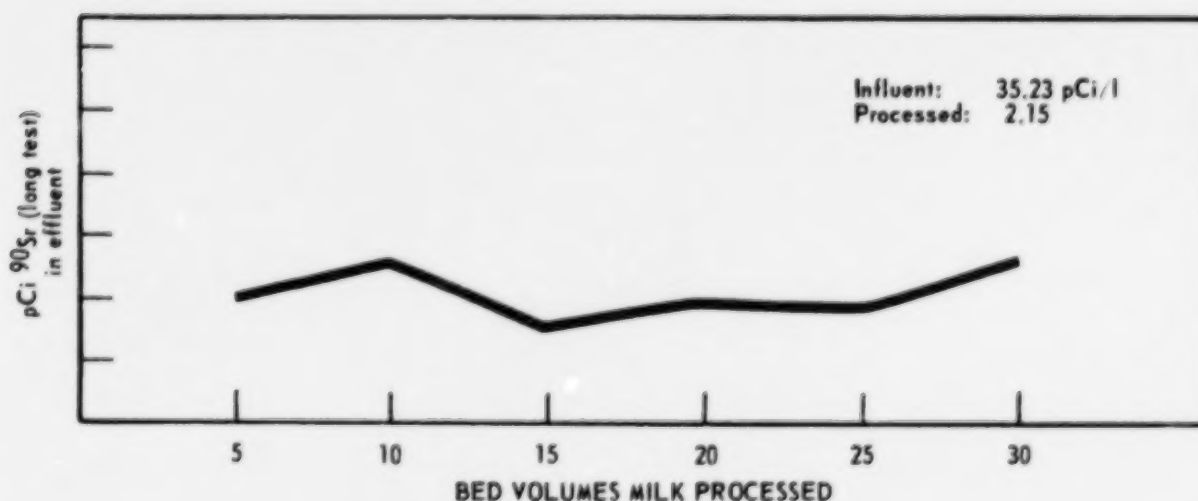


Figure 1. Relation between bed volumes processed and pCi ⁹⁰Sr removed (average values for six runs)

Individual results making up these averages of the six runs ranged from a low of 0.9 picocurie per liter to a high of 6.7 picocuries per liter. It can be seen, therefore, that the removal rate of the process is better than 90 percent, substantiating the results obtained in the pilot plant at Beltsville.

The average picocuries per liter of milk in table 2 is 37.15 which varies from the number given in table 1 because figure 2 represents only five runs compared to the six in table 1. This is a table showing the accountability of strontium 90. The regenerant solution was sampled and assayed for strontium 90. Then calculations were made based on the 14-bed volumes of regenerant to compare the total picocuries of strontium 90 recovered against those that were in the original milk. This table shows that they varied from a low of 78.7 to a high of 137.5 percent recovery. The average figure is 109 percent, which is fairly representative of the accountability of strontium 90. They are not as good as we would like to see because of the problems involved in obtaining a sample of the regenerant, but these results do indicate at least adequate reliability in the sampling and testing procedures.

Table 2. Accountability of strontium 90

Run number	Pounds milk processed	pCi/l milk	Total pCi to column	Gallons regenerant (14 BV)	pCi/l regenerant	Total pCi from column	Percent recovery
143.....	99,340	42.15	1,833,989	5,180	83.53	1,637,689	89.3
176.....	103,880	36.00	1,637,964	5,180	65.72	1,288,506	78.7
233.....	103,540	32.92	1,492,922	5,180	99.58	1,952,365	136.1
246.....	104,260	43.69	1,995,148	5,180	119.35	2,339,976	117.3
260.....	99,840	31.00	1,355,630	5,180	95.10	1,864,531	137.5
Average.....	102,172	37.15	1,663,131	5,180	92.66	1,816,613	109.23

We will now consider results on chemical determinations. In early runs the results obtained on the processed milk were corrected to allow for dilution of the processed milk with water, both the water remaining in the column, at the start of run, and that which is added with the citric acid in the potassium hydroxide was added as approximately a 4-percent solution. Now, when this correction was made there still was an apparent decrease in the protein, fat, lactose, and other values in the processed milk. Determinations were made for these components in the regenerant solution, but nothing was found, so other factors were considered. When the increase in solids in the processed milk, owing to the addition of potassium and citrate ions, was included in the correction calculations, all of the original components in the processed milk could be accounted for.

The results (table 3) demonstrate this point. The protein in the raw milk, for example, was found to be 3.34 percent before processing, and 3.33 percent after processing and after correcting for the dilution

owing to both the water and the potassium citrate. You will note here also that the difference in fat is from 3.77 percent to 3.78 percent, a very slight increase of .01 percent. The lactose content also shows very close agreement, 4.59 percent in the raw milk versus 4.52 percent in the processed milk. Inorganic phosphate averaged 663 parts per million before processing and 671 parts per million, a difference of eight parts per million because of processing. The primary differences, you will note, are in the citrate ion and the potassium ion.

About one-half of the water that gets into the system can be removed by incorporating a small evaporator in the system. About 2 percent of the added water comes from solutions used for adjusting the pH, and about 2 percent comes from the water remaining in the columns after washing the resin. Care must, of course, be exercised during processing to keep added water to a minimum. The principal difference, between the raw and the processed milk from a commercial standpoint, is due to the addition of approximately 0.4 percent of potassium citrate. This is a dilution factor of the original milk solids of about 4 percent.

Table 3 shows that in the three whole milk runs there was an average increase in citrate ions of 2,900 parts per million. This is approximately 0.29 percent, and 1,400 parts per million of potassium, or approximately 0.14 percent. These two add up to 0.43 percent which accounts for the increase of 0.44 percent in total solids.

As Dr. Hazzard pointed out this morning, there is an increase in ash also owing to the increase of potassium. The potassium figure here of 1,400 parts per million compared favorably with the increase of ash of 1,800 parts per million. All of the principal changes in the composition of milk resulting from the processing are largely due to the potassium and citrate ion increase. You notice the curd tension has dropped from 41 to 0, which could be attributed to potassium citrate. There is an increase in specific gravity, a decrease in freezing point, and so forth. There are a number of other minor compositional changes which also can be attributed to the addition of the potassium and the citrate.

Figure 2 shows the variation in the column effluent for the last run made, run 288, for strontium 90, sodium, magnesium, calcium, and potassium. In the case of run 288, strontium 90 shows a slightly higher value at the start of the run. This was attributed to a slight increase in the pH on the column influent. This in turn was due to adjusting the temperature of the milk for the first time to operating at 55° F., and intentionally holding the pH high so as to avoid possible coagula-

Table 3. Comparison of raw and processed milk
(average of three whole milk runs)

Analysis	Raw	Processed ^a	Difference
Protein percent.....	3.34	3.33	-0.01
Ash percent.....	0.73	0.91	+0.18
Curd tension, gram.....	41.0	0	-41.0
Sp. gr. 60° F.....	1.0289	^b 1.0294	+0.005
Freezing point ° C.....	-0.533	^b -0.570	-0.037
pH.....	6.85	^b 6.77	-0.08
Fat percent.....	3.77	3.78	+0.01
Total solids percent.....	12.63	13.07	+0.44
Lactose percent.....	4.59	4.52	-0.07
Citrate percent.....	1,827	4,704	+2,877
Sodium ppm.....	430	411	-19
Potassium ppm.....	1,648	3,014	+1,366
Magnesium ppm.....	156	164	+8
Calcium ppm.....	1,192	1,236	+44
Iron ppm.....	0.28	0.51	+0.23
Chloride ppm.....	1,323	1,419	+96
Inorganic P. ppm.....	663	671	+8
Acidity.....	0.13	^b 0.18	+0.05

$$\text{Correction factor} = \frac{\text{Original total solids}}{\text{Processed total solids} - \text{increase in K and citrate}}$$

Example: Run 260

$$1.078 = \frac{12.89}{12.43 - 0.47}$$

^b Not corrected for dilution effect.

tion in the columns. By the end of the fifth bed volume, it was realized that the 55° F. temperature was satisfactory so the pH was lowered to approximately 5.3 for the balance of the run. It will be noted here that the sodium and magnesium content decreased slightly and the potassium content increased from 2,700 parts per million to approximately 3,000 parts per million during the run. This may be due to minor changes in the pH of the milk caused by changing the addition of the amounts of citric acid. Since the potassium is used to neutralize this, as more citric acid is added, more potassium is required to neutralize it.

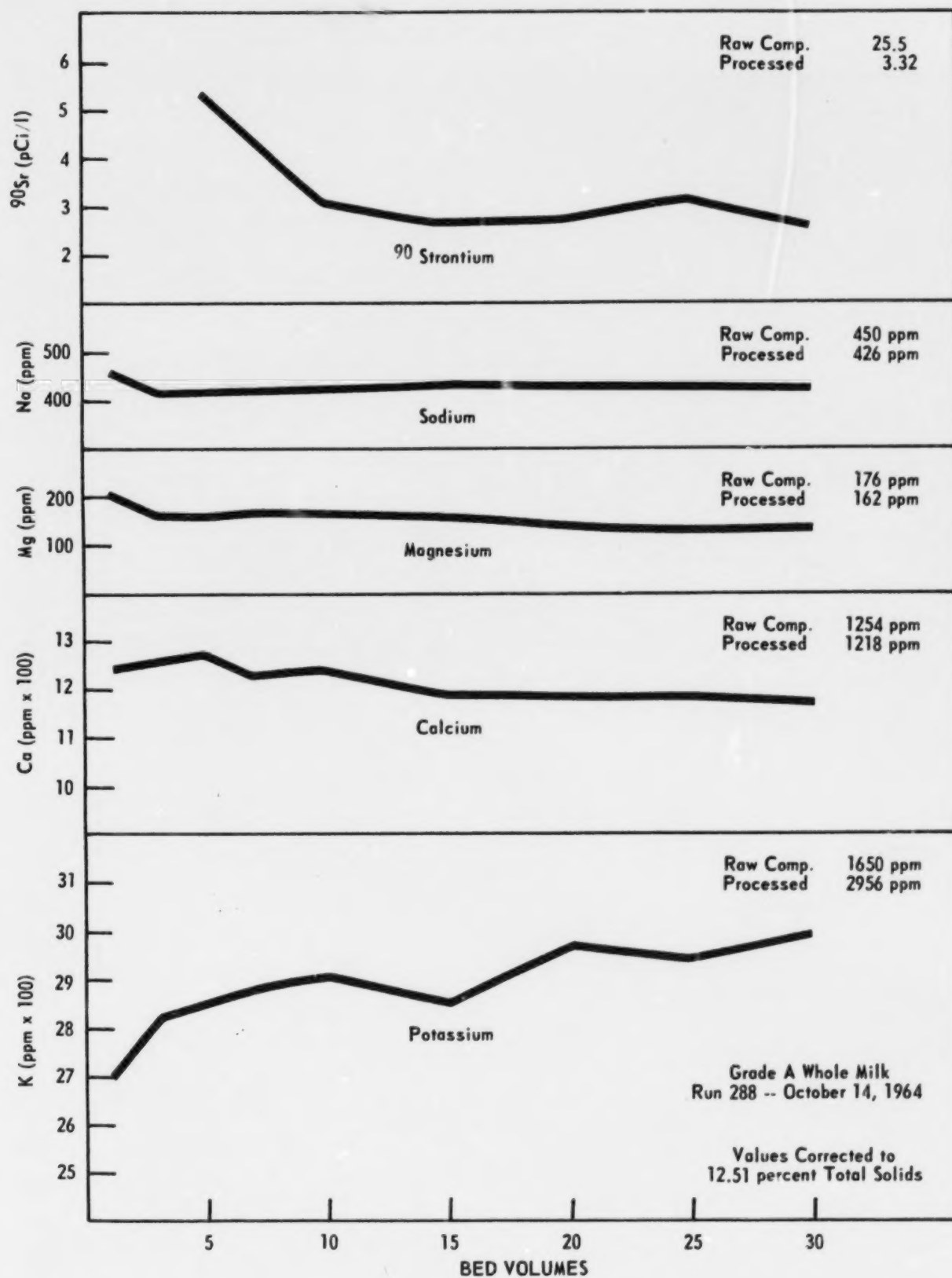


Figure 2. Cation concentration in milk versus bed volumes processed

Whether this is an artifact or not I don't know, but I do know that all of our runs have shown an increase of potassium throughout the run.

Figure 3 shows one run in which we made citric acid and potassium determinations throughout the run, and you will note that there is an increase in citrate as well as increase in potassium throughout the runs.

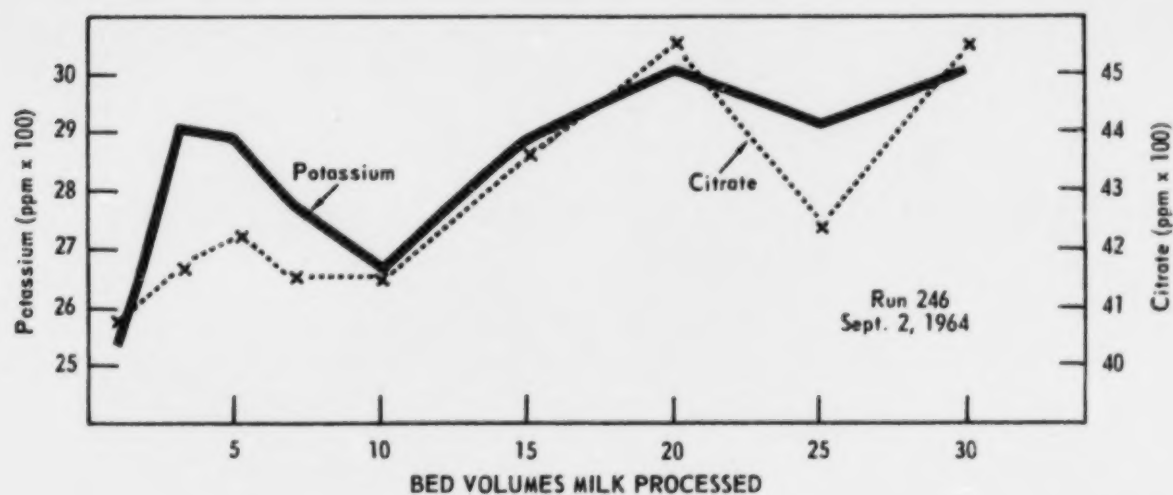


Figure 3. Relation between potassium and citrate contents of milk and bed volumes processed

We will now turn our attention to the bacteriological results. Table 4 shows the bacterial counts on line samples of raw milk taken before and during processing on five runs. These results do not show any increase in count that can be attributed to the operation of the columns. The average of the load counts—this is the raw milk coming into the plant—was 115,000 per milliliter. These are the individual loads, approximately 39,000 pounds for each one, ranging from a low of 38,000 to a high of 250,000 on the five runs represented here. The column effluent at the start of the run ranged from a low of around 30,000 to a high of 580,000. In the middle of the run the column influent ranged from 50,000 to 140,000 and the column effluent ranged from 56,000 to 320,000. At the end of the run the ranges for the influent were from 79,000 to 150,000. As Dr. Read pointed out this morning, there appears to be a slight decrease in counts. On the average, the influent count was 108,000 and the effluent count was 95,200. These results would, at least, suggest that some microorganisms may be removed from the milk during passage through the column at a pH of 5.4. The count on the raw milk, 108,000 versus 115,000, suggests that possibly lowering the milk to a pH of 5.3 may have caused a slight reduction in the standard plate count. We can say at this time that these results indicate that the columns can be operated under satisfactory sanitary conditions.

Table 4. Bacterial counts on line samples of raw milk processed for removal of ⁹⁰ Sr

Sample	Run 176	Run 233	Run 246	Run 260	Run 288
Raw milk:					
Load 1-----	71, 000	90, 000	38, 000	46, 000	250, 000
Load 2-----	120, 000	77, 000	410, 000	98, 000	69, 000
Load 3-----	100, 000	120, 000	110, 000	71, 000	52, 000
Start of run:					
Column influent---	80, 000	50, 000	31, 000	39, 000	580, 000
Column effluent---	33, 000	30, 000	30, 000	39, 000	94, 000
Middle of run:					
Column influent---	66, 000	50, 000	140, 000	68, 000	59, 000
Column effluent---	56, 000	69, 000	320, 000	32, 000	110, 000
End of run:					
Column influent---	120, 000	95, 000	130, 000	41, 000	67, 000
Column effluent---	140, 000	79, 000	110, 000	150, 000	86, 000

Average of load counts: 115,000/ml.

Average of influent counts: 108,000/ml.

Average of effluent counts: 95,200/ml.

Table 5 shows the results of bacterial counts made on the pasteurized processed milk after removal of strontium 90. Milk occasionally had to be pumped from one tank to another in order to obtain representative samples, and because of this unusual procedure high coliform counts were encountered on two of the runs. Once, on run 176, we had a coliform count of 66 per milliliter, and on run 260 we had a coliform count of 117. Run 288—the standard plate counts are less than 3,000 at the start, middle, and end of the run, and the coliform counts are zero all the way through. In general, the bacteriological results indicate satisfactory processing control and equipment sanitation after pasteurization of the processed milk.

Let us now turn our attention to organoleptic changes. In the early runs, owing to problems encountered in the pumping of the milk and in the control of pH to and from the columns, a greenish cast was observed in the processed milk on runs 143 and 176. Also, on these runs, flavor was impaired. Run 143 had a flavor score of 30 (this is the American Dairy Science scorecard system) after processing; run 176 had a flavor score of 37.4 before processing and 35.8 after processing. However, after operating conditions were stabilized, flavor scores were

substantially improved and appearance changes in the milk greatly minimized. On subsequent runs, the color of the milk was almost normal. The greenish cast of the processed milk could only be recognized by direct comparison with the untreated control samples.

Table 5. Bacterial counts on processed pasteurized milk after removal of ^{90}Sr

Sample	Run 176	Run 233	Run 246	Run 260	Run 288
Tank 1:					
SPC/ml.....	3, 000	3, 000	10, 000	3, 000	3, 000
Coliform/ml ^a	2	0	0	117	0
Staph/ml ^b	1	2	1	3	0
Tank 2:					
SPC/ml.....	3, 000	3, 000	3, 600	3, 000	3, 000
Coliform/ml.....	0	0	17	6	0
Staph/ml.....	1	0	1	1	0
Tank 3:					
SPC/ml.....	3, 000	3, 000	3, 000	4, 500	3, 000
Coliform/ml.....	66	0	18	1	0
Staph/ml.....	2	0	0	0	0

^a All colonies appearing on violet red bile agar.

^b All colonies appearing on staph medium 110.

Table 6 shows the results of flavor scores taken on line samples after processing and on the processed composite samples for four runs. The milk was submitted to a trained panel of five to seven members and these are the average scores. As a rule of thumb, for those not familiar with this scoring system, sample scores above 36 are generally not subject to consumer complaints; however, samples with flavor scores of 37 to 40 are considered good to excellent products. Table 6 shows that the average score of the milk before processing was 38.15 and 37.38 after processing. The average decrease in flavor score because of processing was found to be 0.77, and this agrees with one of the speaker's statements this morning regarding results obtained on milk at the Beltsville plant. The difference is not considered large enough to indicate a flavor problem. While not shown on the table, samples of processed milk were held in the refrigerator for several days and again submitted to the flavor panel for flavor evaluation. Since no appreciable change in flavor occurred, the keeping quality properties of the processed milk are considered to be satisfactory.

Table 6. Variation of flavor score with bed volume of milk processed for ⁹⁰Sr removal

Sample	Run 233	Run 246	Run 260	Run 288	Average
Raw composite-----	37.6	38.0	38.4	38.6	38.15
5 BV-----	37.6	35.7	37.6	37.8	37.18
10 BV-----	37.6	36.7	37.6	37.4	37.33
15 BV-----	37.6	36.3	37.6	37.9	37.35
20 BV-----	37.6	37.0	37.8	38.0	37.60
25 BV-----	37.0	36.5	36.4	37.6	36.88
30 BV-----	37.6	37.0	37.8	37.8	37.55
Processed composite-----	37.3	37.0	37.4	37.8	37.38

Average decrease in flavor score because of processing: 0.77.

Let us now consider control procedures for commercial operating conditions. A great many tests were run during the processing of the milk. However, under commercial conditions very few of these would be required. All of the control tests used are capable of being adapted to routine plant operation, and it is not anticipated that problems in the composition control of the effluent milk would be encountered. The pH checks by the laboratory are essential and must be done at regular intervals during processing in order to insure the satisfactory operation of the columns. The titratable acidity, which is a measure of the potassium citrate increase, can be used as a very quick check on operating conditions. When we speak of acidity in the dairy industry, we sometimes think of developed acidity, but there is also acidity in the milk which is due to the buffer capacity of the milk. When you increase the buffer capacity by adding potassium citrate, you increase the titratable acidity. This is a very quick and useful tool which can be used to guide the process as it is going along. On runs 143 and 176, for example, which I mentioned were not very satisfactory from an organoleptic standpoint; the titratable acidity varied from 0.185 to 0.27 which is enough to give the milk a sharp, acid taste, as Homer Walter mentioned. However, on the remaining four runs where the process was stabilized, the titratable acidity ranged from 0.13 to 0.17 percent and generally increased at a steady rate in the same way that potassium and citrate ions increased at a steady rate.

Another requirement for proper control of the processing would be that total solids determinations be made so that the product can be standardized after processing. The water added to the milk can be evaporated on a continuous basis so that total solids tests would be necessary to be certain that the amount of water removed was equal

to the amount of water that was put in during the processing. Color and flavor tests should also be made on each batch of milk processed. Strontium 90 tests can be made by the short method and the results applied as required to both the raw and the processed milk and these made available within a relatively short time.

In summary then, the data we have obtained indicate that the commercial operation of the fixed-bed, ion exchange system is capable of removing more than 90 percent of the environmental level of strontium 90. There is no increase in microbial population, and a very slight decrease in flavor score. Except for the added potassium citrate, there is only a very minor change in composition. In the event that environmental contamination should exceed safe tolerances, this process appears to be a commercially feasible and practical means for substantially reducing the levels of strontium 90 in milk.

DISCUSSION

Question:

I am concerned about running these columns for 8 hours at 55° F. I am surprised that you didn't get any increased bacterial activity. Did you do any psychrophile counts?

Answer:

No, these are standard plates, incubated at 32° C.

Question:

I would like to have someone answer why run an 8-hour process—you could get just as much strontium out in 4 hours. This is a better process, microbiologically, so far as I can see. I can't see any reason for running 8 hours, when you could do it in 4. Would someone like to comment on this?

Do you want to say anything, Dr. Edmondson?

Answer:

Well, I mentioned the possibility of running 8 or 4 hour operations—that is, faster bed volume flow rate and the removal is not much—in one case you switch after 4 hours and in the other case after 8 hours. In the 4-hour operation you have two columns to maintain an 8-hour continuous run. In the 8-hour run you only have to have one column. There is a little more flexibility in some respects

where you run split lots of milk, or just for a short run in a 4-hour setup, than there would be for the other. However, on switching columns there is the considerable problem of the dilution effect where you get the column full of water head space, and the more often you have to switch, say for the 4-hour run, the greater this problem of dilution would probably be. So, the 8-hour run in that respect will have an advantage. The total amount of regenerant required should be about the same. With a 4-hour run, if we have four regenerant tanks, they would only be one-half the size. That might be an advantage as far as the 4-hour run is concerned. The total amount of salt solution required for regeneration should be about the same for each volume of milk processed. Because of this business of keeping an operation going without having to interrupt and make the switches necessary for water dilution and cleanup of column, I favor the 8-hour run unless most of this has to be done during a normal working day within an 8 a.m. to 4 p.m. operation. Most plants don't seem to pay much attention to that, however.

RELATIONSHIP OF THE ION EXCHANGE PROCESS TO PRESENT DAY COMMERCIAL FLUID MILK PROCESSING OPERATIONS

*E. L. Ruppert*¹

Mr. Ruppert:

We have attempted to put down a few factors which will pertain to the relationship of this process to present-day, commercial fluid milk operations. These points have already been discussed in some detail, so this will really be in the nature of a summary of these factors.

A process has been developed that effectively removes cationic radio-nuclides from milk. This process has been evaluated in the laboratory, in a pilot-plant scale operation, and now in an installation of commercial magnitude.

The relationship of this process to present-day commercial processing of fluid milk now becomes a factor of significant concern to the government agencies involved, to the dairy industry, and to the public.

We will probably all be questioned soon regarding when and how this process should be used.

This process was conceived and has been developed as a process for "emergency" use. The "emergency" was considered to be such time as levels of specific radioactive materials should present a potentially significant public health hazard. The early public announcements in December 1961 regarding the pilot-plant evaluations of the process emphasized this "standby" philosophy as follows:

"Development of the ion exchange process for removing radioactive strontium from milk was undertaken as a standby measure 2 years ago

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in a cooperative project participated in by the Public Health Service, the Department of Agriculture, and the Atomic Energy Commission. It was believed that such a process would become important in the event of a nuclear attack. In addition, it might be used in case of an accident in an atomic energy installation resulting in the release of large amounts of radioactivity, or a vastly increased contamination of the atmosphere as the result of a nuclear testing program of far greater magnitude than any country has undertaken to date."

The nutritional studies that were performed along with the pilot-plant evaluations were aimed at determining any gross changes in the nutritional value of the milk. Tests were run on one generation of rats and pigs; no detrimental effects were observed during these studies. However, to determine the possibility of long-range effects, studies of animals for longer periods of time would be advisable. Thus, while we have a general judgment regarding the nutritional acceptability of the product of this process for short term or emergency use, we do not know whether it will be satisfactory for extended or prolonged use.

The process has been operated by the plant personnel, under the supervision of professionally and technically oriented people, for the various test runs. Milk plant personnel would need adequate training and supervision before operating the equipment.

The studies of microbiological safety and the cleaning and operating procedures have been directed largely toward specifying a procedure which seems to be satisfactory for the cation column. This method may not be the optimal or most desirable method. Operational experience with the system may result in other methods of cleaning and sanitizing the equipment that are more satisfactory. Time and funds have not permitted a thorough microbiological study of the full-scale system. Such studies should be made to delineate any problems which may be inherent in such a full-scale system. It is doubtful that a control agency, operating a Grade A milk program, would be willing to accept the process in its present stage of development for routine use without these further studies. As with any other milk process, it should be under the control of the appropriate regulatory health agency, since it is entirely possible to significantly increase the total bacterial count of raw milk if the process is not operated properly. Under such conditions, enterotoxin-producing staphylococci could multiply and produce toxins in the resin column, and thus contaminate the milk. Both of these situations would be potentially dangerous to the milk consumer. Recent studies indicate that even dead bacterial

cells may produce gastroenteritis when consumed in large quantities. The dangers of the heat-stable staphylococcal enterotoxins are well known.

There are other factors that must be considered. This process will no doubt increase the cost of milk processing, and thus may have some bearing on the nutritional status of a part of the population—particularly if this commodity is priced out of their reach.

Still another question relates to the advisability of installing ion exchange process equipment so that it will be available when and if an emergency occurs that would warrant use of the process.

The term "emergency" is not, at this time, sufficiently defined to delineate when and how long such a process should be used to treat a given milk supply.

In summary, we have a process that could be used when and if it is necessary for protection against cationic radionuclides. There are some potential microbiological risks involved in the widespread use of the process which still need to be studied and which must be evaluated in relation to emergency use. If practical, other protective measures, such as immediate removal of dairy cattle from fresh forage and importation of milk from uncontaminated areas, would have to be considered under such a defined emergency. It is also possible that improved processes and equipment may be developed in the near future which will be more effective. At the present time, considering these factors, we would not encourage the use of this equipment as a part of a routine milk processing operation.

ADDITIONAL RESEARCH AND DEVELOPMENT ACTIVITIES CONSIDERED FOR TREATMENT PROCESSES

*R. E. Bales*¹

Mr. Bales:

The system that has been under discussion throughout the day has been the full-scale, fixed-bed process. Hopefully, we will successfully culminate several years of development and testing in the next several days with the final 16-hour milk run. Concurrent with the development and testing of this full-scale process, a number of other processes have been considered, and some of them are still under consideration. I would like to discuss briefly the research and development activities concerning these treatment processes that are worthy of note at this time.

FIXED-BED-RELATED STUDIES

These include the consideration of testing a full-scale, fixed-bed system which includes both the anion and cation processes. To date, the full-scale tests have considered only the removal of cations, and we have purposely omitted anion work as a part of these tests rather than introduce new parameters and problems which may be related to the anion process.

The capabilities of a combined system certainly must be considered. Also, we would like to consider a test of the full-scale system under what we might term "field conditions." This does not reflect upon our friends at Producers Creamery, but they are certainly experienced as are no others in the world, and we would like to consider testing under conditions a little closer to an actual field trial.

¹ Research Branch, Division of Radiological Health, Public Health Service, U.S. Department of Health, Education, and Welfare. Present assignment: Chief, Protective Action Section, Environmental Surveillance Control Program, National Center for Radiological Health, Rockville, Md. 20852.

MOVING-BED SYSTEMS

The moving-bed, ion exchange system was mentioned earlier today by Dr. Edmondson, and as he pointed out investigations were started sometime ago, and he described the bench-scale system studies at Beltsville. This unit was about 20 feet high and was constructed of 1-inch-diameter glass. The moving-bed, ion exchange system is not new by any means. It has been used for a number of years in the chemical processing industry, in metallurgical separations, and is being used in full-scale water treatment for municipal supplies. However, its application to the processing of milk has been limited to bench-scale testing. Therefore, research and development work has continued. At Cincinnati, Dr. Murthy has continued work on milk chemistry, and Dr. Read has continued to be concerned with the solution of problems associated with sanitation. The work to date indicates that the sanitation problems are formidable. The Department of Agriculture and the Public Health Service have jointly initiated a research and development contract with the Chemical Separations Corp., Oak Ridge, Tenn., for the fabrication of a pilot-scale, moving-bed unit which will have a capacity of about 100 gallons per hour. This is being fabricated of stainless steel and glass and will incorporate a double loop providing an anion and cation removal system. We also have a small bench scale unit under construction which will be used in conjunction with the testing of the pilot-scale unit at Montgomery, Ala.

Since the moving-bed system is also an ion exchange process, many of the considerations that have been discussed today relative to the fixed-bed process are also applicable to the moving-bed process. The gross difference is the intermittent movement of the resin bed through a loop system. At the same time, I should point out that there are many new problems associated with the development of a large-scale, moving-bed, milk-treatment system. Considerable development and work remains to be done before we can produce a commercially feasible system as complete as the fixed-bed cation process is at this time.

Figure 1 is an isometric sketch which shows the general piping arrangement of a folded loop. This configuration was developed in order to compress this system. Rather than have a 20-foot vertical column, we have asked the contractor to fold the loop over so that it will fit into a module which, along with all of the processing equipment for the 100-gallon per hour unit, can be accommodated within a standard 40-foot trailer truck.

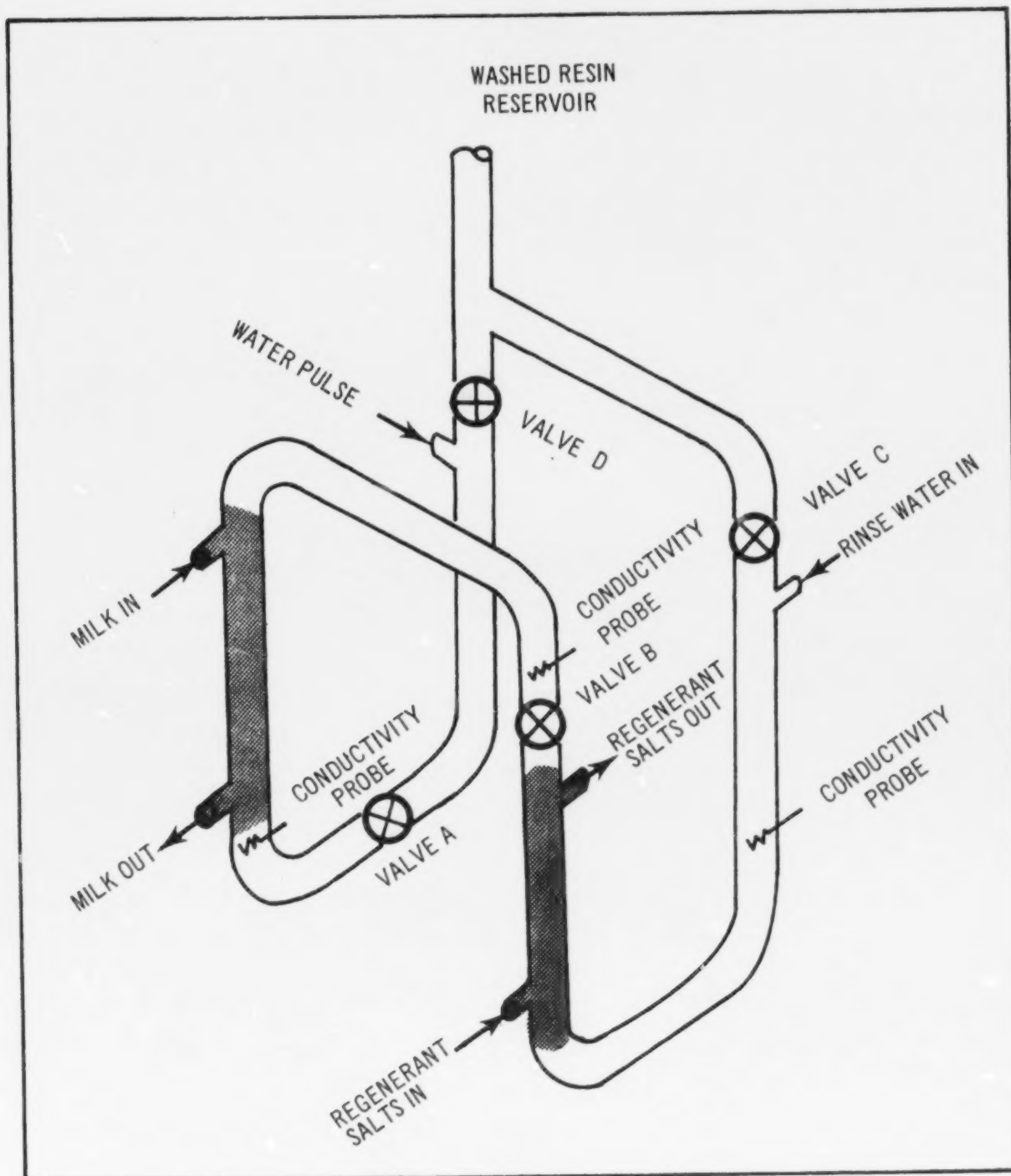


Figure 1. Moving bed ion exchange system folded loop arrangement for milk decontamination pilot plant

The sketch shows how the materials flow through this system. This is a cation loop. An anion loop would be similar, since the general ideas are the same. Milk flows in, flows down, and the resin will flow countercurrent to the milk flow. The resin bed is moved by an intermittent water pulse applied for a period of several seconds. This water will pulse only when the valves marked A, B, and C are open. When valves A, B, and C close, valve D opens.

When the water pulses, the resin moves a few inches around the loop. The milk-water interfaces, as well as the water-regenerant salt interfaces, also move ahead a few inches in the column. There are points in the system, as shown, where conductivity probes detect the passage of an interface, and these sensors actuate appropriate valves, including a water inlet valve, not shown. The activated valves then allow the pressure differentials to react on this system so that the interfaces then move back to the sensors. This means there is a riding milk column which moves ahead a few inches with the resin bed, and then moves back relatively slowly after the pulse. In the same way, in the regenerant section the regenerant salt solution and water interface moves back until the sensor activates the water line valve to close.

The last section also washes the regenerated resin. The following pulse moves the regenerated and used resin past valve C into a resin reservoir leg. When valve D opens, the clean, fresh, regenerated resin drops down into the leg below, thus completing the cycle. This then becomes a continuous process, since milk continues to flow at all times through the milk section.

This 100-gallon-per-hour pilot plant will be arranged so that it will fit into a 40-foot semitrailer truck. It will be designed to operate directly in the trailer alongside a milk plant. The system will be constructed within a frame which can be removed from the truck, if necessary, and dropped to the ground. The frame will be fitted with wheels so that the unit can be suitably positioned if on a good, flat surface.

ANION TREATMENT PROCESSES

The use of anion exchange resins has been referred to several times today although we have been discussing the strontium removal processes. The anion removal system is similar to the cation system in that, by ion exchange, it is possible to remove unwanted anions. In a similar manner, we also remove some of the desirable anions from milk. These then, must be charged onto the anion resin to replace those removed to retain the proper anionic balance in milk. The anion of interest in such a removal process, in all likelihood, would be radioiodine. It has been demonstrated that anion resins successfully remove radioiodine from milk. Further investigation of anion resin efficiency and stability are underway at Cincinnati, Ohio, and Montgomery, Ala.

It has been pointed out today that potassium is necessarily added to milk when we neutralize with potassium hydroxide. Since milk normally contains potassium, this additional potassium may be of minimal concern as indicated by nutritional testing. However, with the anion process, it may be possible to charge an anion resin with hydroxide ions and use this processing stage for neutralization of the cation treated milk, eliminating the addition of potassium. This is another consideration which will be tested with the combination anion-cation system.

ELECTRODIALYSIS PROCESS

Again, this is a process that has been proven in large-scale, ion removal installations, notably for water desalinization and also in the desalting of whey. It has *not* been proven for large-scale milk processing. Very simply, this is a process for removing ions from solutions by the use of membranes which allows passage of ions according to the electric charge on the ions and the charge on the membranes. There is essentially no flow of the fluids themselves through the membranes. In this process, an electrical potential is maintained across a number of the cells. The cations move in one direction and the anions in another, through respective membranes.

The schematic drawing (fig. 2) shows a unit cell as used in a system for the desalinization of water. Here, essentially, we have two streams of water separated by a membrane. One stream of water will come out of this system with a lower concentration of salts, and the receiving stream (the other stream) will come out of the system with a higher concentration of salts. The schematic drawing (fig. 3) shows a unit cell as would be applied to a milk system. This particular system has been tested on a very small scale by Ionics, Inc., under an Office of Civil Defense contract. In this case, anion and cation membranes are used in the sequence shown for the unit cell. The anions will move as shown through the anion membranes and the cations will move in the opposite direction. In the milk system, the ions migrate from the milk stream into the adjacent aqueous stream, and adjacent salt solution allows ions in proper proportions to migrate into the milk, replacing the desirable salts which are being removed with the radionuclides. Each of these ions migrates at a different speed through a given membrane. It happens that cesium moves very rapidly so that cesium removal efficiency is especially high with this kind of system. Strontium is removed with good efficiency. The rate of ion migration and the efficiency of removal is dependent upon the potential that is

placed across these cells and upon the number of unit cells that are stacked. In a processing plant, these membranes and separate sheets are stacked by the hundreds. They are prepunched so that the proper solutions will flow between the proper anion and cation membranes, and the various solutions then can be pumped into and out of the system allowing for recycling, makeup, and discharge as appropriate.

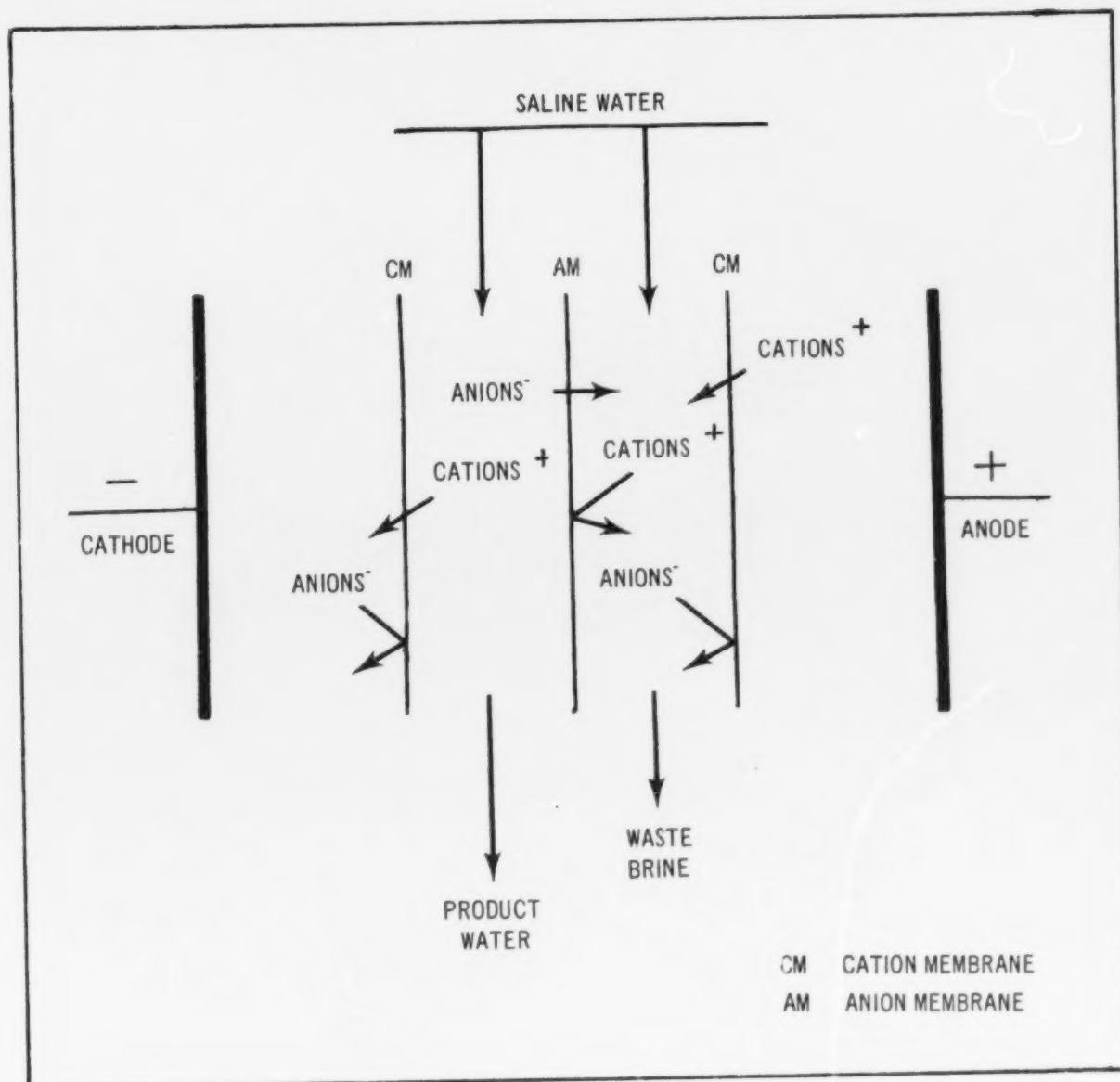


Figure 2. Unit electrodialysis cell as applied to water desalinization

There is a limitation to the voltage that can be applied. Otherwise, there would be no limit to the capacity for removal. However, a point is reached for a given system where, as the potential is raised, polarization occurs at the surface of the membrane, and the voltage will rise because the water will ionize and there will be little or no removal of the salt ions. Thus, there is a threshold voltage below which these units must be operated for any particular system. This then deter-

mines the capacity for the unit cell and determines the number of cells that are required for a given total capacity. This system has considerable merit and certainly should be considered for further research and development in the future.

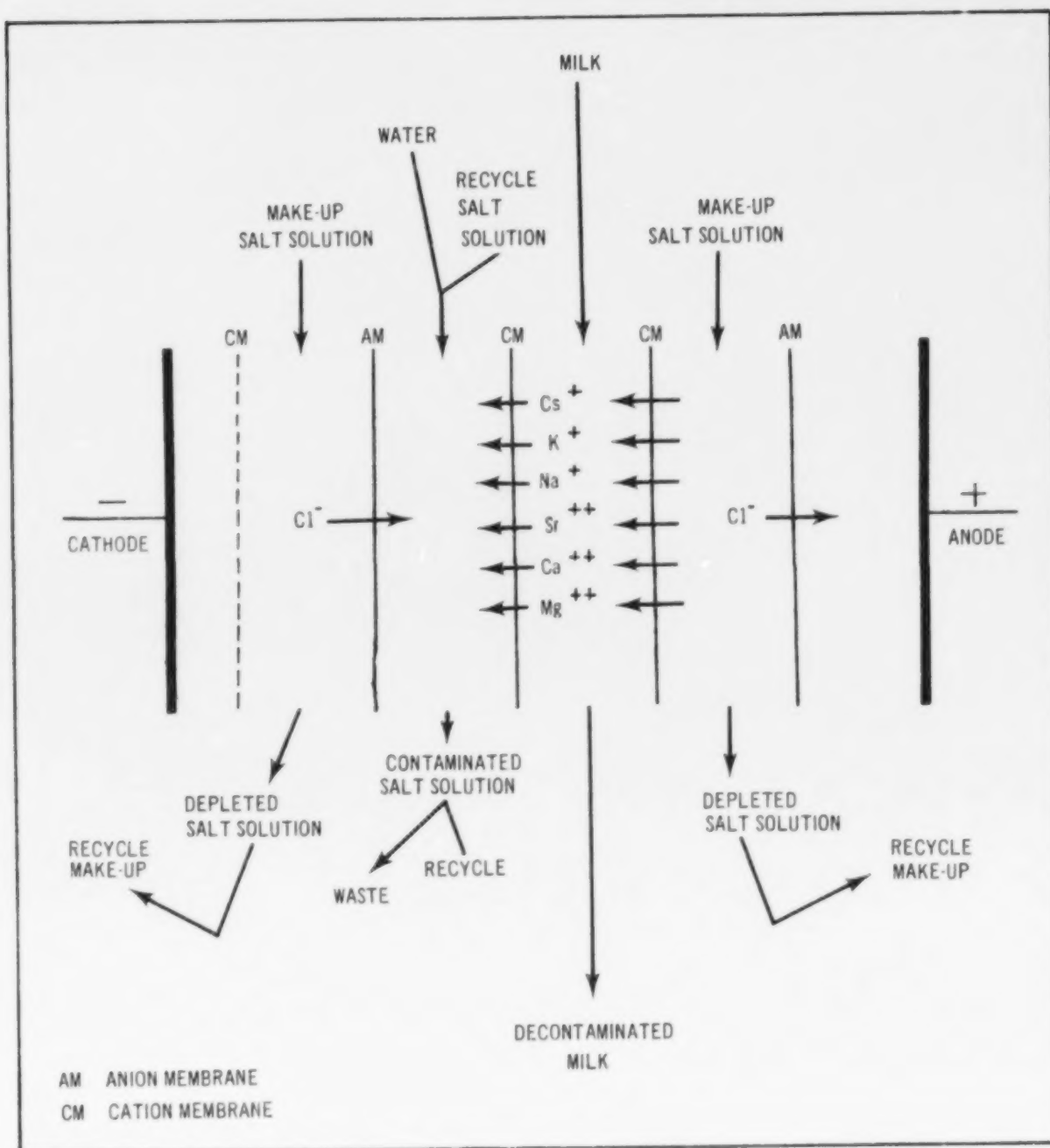


Figure 3. Unit electro dialysis cell as applicable to milk decontamination

COPRECIPITATION PROCESS

This method has received considerable publicity since it appears to be effective and simple. A method suggested by Silverman several years ago consisted of adding calcium phosphate to milk, and allowing

a precipitate to form which coprecipitates the strontium with calcium phosphate, and thus effects an appreciable removal of the radio-strontium. Dr. Edmondson and his coworkers studied this method and found that the coprecipitate removes a large amount of the calcium, as well as other salts and nitrogen from the milk, thus excessively affecting the quality of the milk. This method was therefore not pursued any further.

CONCLUSION

Considerable research and development work has been done on ion removal systems as applied to aqueous media for a variety of purposes. Several of these systems appear to be attractive for application in the removal of radionuclides from milk and other foods. Especially suited to the processing of fluid milk are the ion exchange and the electro-dialysis processes. In addition to the full-scale, fixed-bed cation exchange process development, pilot-scale development is in progress for the moving-bed and cation-anion exchange system, and preliminary bench scale work on dialysis is contemplated.

SUMMARY

Dr. Todd:

I have asked Mr. Terrill to summarize the conference.

Mr. Terrill:

You are not only a good audience, but most of you are a captive audience.²

I wish to emphasize, particularly to people who were late getting here, that this meeting was held in an attempt to familiarize public health people, industry people, and agricultural people with the progress we have been making in this process development.

If there are any ways in which we can further stimulate thought in this general area on the part of any group, we want to do so. I think our technical people have given you much information in this short period to illustrate what we know and where we think we are going. As was pointed out this morning, there are many valued judgments involved just in terms of the definition of urgency—when you dig into the strontium problem you find that this is going to be the basis of judgment. As to the overall nutritional effects, any change will in-

² Because of a record snow storm on the previous evening, many attendees were detained en route and in departure.

volve not only the nutritional aspects per unit of intake but also the total quantity. If any of you have any further suggestions, not only up to now, but after looking at the plant, as to the direction we should take, or if you have any questions please feel free to deal directly with our technical people. Certainly, I don't want to speak for Agriculture, but on the basis of my experience I would say generally you can deal with them and get any information that your people need to evaluate this process. I hope that all of you who participated in this evaluation will make comments and suggestions to the people who can serve in the best interest of all of us to produce a satisfactory process. I think that is all I have to say, and I thank you for being such a good audience.

Before we close the meeting, I would like to take this opportunity to acknowledge some individuals whose contributions have helped to make the meeting a success. The first one is Mr. Munzer, who so ably projected the slides that we have had very few, if any, pauses. The last several meetings I have attended, we have had problems with the projector and other paraphernalia, and I think the meeting went very smoothly because of John Munzer's efforts. The young lady in the back there, Mrs. Smith, is Mr. Munzer's secretary, and I think she is doing a good job of recording the proceedings.

APPENDIX I

PERTINENT ARTICLES AND PAPERS (BIBLIOGRAPHIC MATERIAL)

The following documents were contained in kits distributed to participants:

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APPENDIX II

TASTE TEST TABULATIONS

**Table 1. Average taste panel flavor scores for whole milk
(samples 3 days old)**

	Control sample	Processed sample
February 21, 1965.....	37.0	37.0
February 25, 1965 (first run).....	36.9	63.9
February 25, 1965 (second run).....	37.1	36.0

**Table 2. Taste test results for whole milk processed on February 21, 1965
Symposium attendees—blind taste test on February 24, 1965**

	Number of persons	Vote
Preferring treated milk.....	19	A
Preferring regular market milk.....	26	B
No preference indicated.....	9	O

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